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ACIDIC PRECIPITATION IN ONTARIO STUDY

AN ANALYSIS OF THE EFFECTS OF THE SUDBURY EMISSION SOURCES ON WET AND DRY DEPOSITION IN ONTARIO

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SYNOPSIS

During the period of June 1982 to March 1983, there was an extensive shutdown of the smelters in Sudbury. This report is written to address two questions:

1. Is the impact of the smelter shutdown on wet and dry deposition observable?
2. Can the smelter impact be quantified?

Monitoring results from the Acidic Precipitation in Ontario Study (APIOS) event and cumulative deposition networks during July to March of 1980-81, 81-82, and 82-83 were examined.

Due to the lower time resolution of the cumulative data and the large meteorological variability in the data, comparison of the overall averaged results of the operating and shutdown periods obtained within a radius of 250 km of the smelters failed to yield any evidence of systematic and definitive changes in average precipitation concentration due to the smelter shutdown, except for copper. Though not quantitative, a comparison of precipitation concentration contours obtained over the operating and shutdown periods indicated evidence of a smelter impact on $SO_4^=$ and Cu. However, the comparison of the corresponding deposition contours did not.

By classifying the event data by their synoptic meteorological and air back trajectory patterns, the potential smelter contribution to wet and dry deposition was quantitatively assessed, and is summarized in the following table according to site location and parameter of interest. The values shown in this table are thought to be near the upper limits of the possible smelter contribution (i.e. the actual contribution is less than these values).

Site	Parameter	Sudbury Smelter Contribution to Wet Deposition (% of total)	Sudbury Smelter Contribution to Dry Deposition (% of total)
Dorset	$SO_4^=$	11 - 12%	18 - 28%
	SO_2	**	31 - 37%
Railton	$SO_4^=$	2%	*
	SO_2	**	27%
Longwoods	$SO_4^=$	*	*
	SO_2	**	5%
Fernberg	$SO_4^=$	*	*
	SO_2	**	4%
Kapuskasing	$SO_4^=$	*	46%
	SO_2	**	47%
Chalk River	$SO_4^=$	12%	17 - 20%
	SO_2	**	25 - 26%

* Not estimated due to limited data.

** Not measured in precipitation samples.

It should be noted that the stratified analysis was based on only a limited number of samples, therefore the estimated percentage contribution to wet and dry deposition due to the Sudbury smelters should be considered as tentative and should be confirmed when future operating and shutdown data become available.

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Our thanks are also due to Ontario Hydro and the Atmospheric Environment Service, for making available their air and precipitation data for Kapuskasing and Chalk River respectively.

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1. INTRODUCTION

Sudbury is an area of major environmental interest because it is the location of two smelting/mining companies, namely, INCO Limited and Falconbridge Limited. Both smelters emit large quantities of SO_2, which accounted for 45% and 7% respectively of the Ontario SO_2 emissions in 1981 (Yap, 1984).

During the period of June 1982 to March 1983, there was an extensive shutdown of the smelters in Sudbury. Even though during parts of this period, there were SO_2 emissions associated with the Falconbridge operation (June 1 to 26, 1982 and January to March 1983), these emissions were considered to be small relative to those of the regular total Sudbury Smelter emissions. In order to relate receptor observations to source emissions in the acid rain debate, it is pertinent to examine the empirical data to address the following two questions:

1. Is there any observable effect on wet and dry acid deposition as a result of the Sudbury smelter shutdown?
2. Can the extent of impact be quantified geographically?

This report is a summary of some attempts made to answer these questions using the wet and dry deposition data from the Acidic Precipitation in Ontario Study (APIOS) networks, Environment Canada's Air and Precipitation network (APN) and Ontario Hydro network.

2. DEPOSITION NETWORKS AND DATA

The Air Resources Branch of the Ontario Ministry of the Environment has been quite active in the monitoring of precipitation quality in the Sudbury area since the late 70's (Chan, 1982). It was recognized then that acid rain was not a problem of a local origin, but rather it was related to long range transport of air pollutants. Hence province-wide networks were established in 1980 to monitor both wet and dry deposition.

There are two networks operated by the Air Resources Branch in the province of Ontario: a cumulative network which samples on a 28 day basis (this network sampled on a monthly basis from network inception till January 5, 1982) and an event network which samples on a daily basis. In both networks, wet deposition is directly measured and dry deposition is inferred from ambient air concentration measurements. The primary function of the cumulative and event networks is respectively to quantify the long term deposition pattern and to determine the origin of the pollutants being measured. At this moment, the cumulative network has 36 wet monitoring stations of which 23 are equipped with air measurement instrumentation. The event network consists of 15 wet deposition sites and 4 of them also sample air on a daily basis. The spatial distribution of the stations of which results are being used in this analysis is shown in Figures 2-1 and 2-2 and details of the networks are documented elsewhere (Chan et. al, 1982 and 1984).

Chemical analyses of the collected samples are carried out in the Laboratory Services and Applied Research Branch (LSARB). Laboratory and field data are merged and stored in the Ministry's Sample Information System (SIS). The laboratory data are further subject to stringent validation/screening criteria such as sample integrity checks (e.g. ionic balance, theoretical vs observed pH/conductance, free hydrogen vs total hydrogen ions etc), gross limit checks and Dixon Ratio test (Kirk 1983 and 1984). Outliers are flagged but not deleted and are retained in the data base together with the validated data.

3. DATA ANALYSIS

The analysis performed here places a higher emphasis on the wet deposition results because there are more wet deposition data available than dry deposition/air concentration data.

In order to fully understand the relationship between source and receptor, information on source emissions, intermediate processes such as dispersion/transport, chemical transformation, and deposition at the receptor must be available. One difficulty often encountered in source impact analysis is the large variation in the data due to meteorology. This point can be illustrated by the outcome of the gross averaging of the observed results for periods corresponding to various seasons and source emission configurations, and is the theme in Section 4.

An examination of the gradient of concentration/deposition as a function of distance from the source yields qualitative information on the potential impact of the Sudbury sources. Especially by taking ratios of data to a tracer such as NO_3^- which is not emitted by the smelters, the variability due to meteorology is partially accounted for. This approach is described in Section 5, using the cumulative wet deposition data.

One approach of examining the source/receptor relationship is by mathematical modeling, holding the meteorological component constant but adjusting the emissions factor (Ellenton and Misra 1984). An alternative approach is to group the results under similar meteorology and compare different emission configurations. A first-cut attempt at this is by simple air parcel trajectory analysis (Kurtz and Yap, 1984).

Due to the uncertainties associated with simple air parcel trajectory groupings, data need to be examined further with a view to the concurrent synoptic pattern, in order to estimate quantitatively the extent of the impact by the point source (in this case Sudbury) on a receptor (in this case monitoring site such as Dorset). Sections 6 & 7 summarize results using this detailed stratification scheme to estimate the impact on wet and dry deposition of the Sudbury source on distant sites.

4. STATISTICAL ANALYSIS TO EVALUATE SUDBURY SHUTDOWN IMPACT USING CUMULATIVE NETWORK DATA

4.1 Analysis:

Results obtained at stations from the APIOS cumulative network within a radius of 250 km from Sudbury are grouped together on a seasonal basis (e.g. January to March, April to June, July to September and October to December and these periods are labelled in the figures as 02, 05, 08, 11 respectively).

Table 4-1 summarizes the observation periods, number of sample points used, precipitation-weighted mean concentrations (average concentration weighted by the precipitation depth) and the number of missing data points in this calculation. Of particular interest are the 3 shutdown periods (8208, 8211 and 8302) and their comparisons with respect to the preceeding operating periods. The results for selected parameters are represented in Figures 4-1 to 4-5 for easy visual comparison. The reason for grouping data in the reported fashion is to allow a comparison for the same seasons, and thus decrease variability due to seasonality. Table 4-2 shows whether or not there was a statistically significant decrease in concentrations during the shutdown period.

4.1.1. H_f^{+} (free hydrogen ion): Figure 4-1

Seasonality is noted in the data; highest values occur in the summer or in the fall. The effect associated with the smelters' shutdown on H_f^+ is somewhat irregular; H_f^+ decreased in the 08 period but increased in the 11 and 02 periods. The observation may reflect an irregular neutralization pattern due to the presence of alkaline materials (e.g. NH_4^+, Ca^{++}, etc.), suggesting that H_f^+ is perhaps not a good parameter to be used for source impact evaluation.

4.1.2. H_t^+ (total hydrogen ion): Figure 4-2

The trend in H_t^+ is again quite irregular for the same reason given above for H_f^+ and therefore should not be used as a definitive indicator of smelter impact.

4.1.3. $SO_4^=$: Figure 4-3

Except in the case of the 08 period where a consistent decrease was observed during the shutdown with respect to the operating periods, both the 11 and 02 periods exhibit a high-low-high pattern which is not easily interpretable.

4.1.4. $N\text{-}NO_3^-$: Figure 4-4

Seasonality is again observed of this parameter. Even though there is a mild variation for each of the seasons (08, 11, 02) between the shutdown and operating periods, the change is not statistically significant, being consistent with the fact that only negligible emissions of NO_x originate from the Sudbury smelters.

4.1.5. Cu: Figure 4-5

Cu is a parameter characteristic of smelter operation. In the display of the average seasonal concentration, there is a consistent decrease showing the effect of smelter shutdown on copper concentrations even though the decrease is not always statistically significant.

4.2. Summary

An attempt was made to examine whether the effect, due to the shutdown of the Sudbury smelters on precipitation quality, is observable. The grouping of data from stations within a 250 km radius of Sudbury suggests no systematic, statistically significant decrease during the shutdown period, except for Cu. This points to the fact that because of the large variability in the results associated with meteorology and other factors, more sophisticated approaches such as those using stratification schemes have to be used.

5. IMPACT OF SUDBURY EMISSIONS ON WET DEPOSITION IN ONTARIO

5.1 Approach

This analysis attempt employs wet deposition data from the APIOS cumulative sampling network. Precipitation depth weighted concentration and wet deposition contours of selected parameters were calculated for both the shutdown period and the corresponding operating periods in the preceeding years. The period selected is from July to March because of data availability. Only stations with at least five (for 80/81 and 81/82 periods) and six (for 82/83 period) data points respectively were retained in the contour calculations. The calculation of the weighted concentration is straightforward, by using the available data on precipitation gauge depths and observed concentrations. Deposition was calculated by multiplying the weighted concentration with the corresponding total precipitation depth for the period of interest. Because of the complexity in the interpretation of deposition data (which depends not only on concentration, but also on precipitation amount, which can be quite variable), more emphasis will be placed on the discussion of the concentration contours.

5.2. Concentration Contours

Plots for H_f^+ (free hydrogen ion), $SO_4^=$, N-NO_3^- and Cu are discussed below. Due to the fact that the free hydrogen ion concentration is influenced by potential neutralization by alkaline materials (e.g. NH_4^+, Ca^{++}, Mg^{++}) which may be both of local or long distance origins, these plots do not display readily recognizable patterns from which definitive conclusions regarding emissions impact can be drawn.

The following summarizes patterns for the three July to March periods of 1980/81, 81/82, and 82/83 of $SO_4^=$, NO_3^-, and Cu. Missing data are replaced by "-9.00" in the figures. Station identification is summarized in

Table 5-1. Aside from the regular APIOS data, results obtained at stations near Sudbury (#37 Burwash, #38 Lively and #39 Hammer) which are retained from the Sudbury Environmental Study are also used in the contours production.

5.2.1. $SO_4^=$

Figures 5-1 to 5-3 are precipitation depth weighted concentration contours for the three periods of interest. Common features are:

(i) there is a general S to N concentration gradient, decreasing from S to N.

(ii) there is an elevation around the vicinity of Sudbury - the elevation during the shutdown period seems to be less pronounced with respect to that of the operating periods and may be attributed to possible effects of local dust from the tailings areas near the smelters.

Both these facts together qualitatively point to the effect of the Sudbury emissions on $SO_4^=$ precipitation concentration. To show the relative changes during the shutdown period with respect to the operating periods, ratio plots (Figures 5-4 and 5-5) are generated. Except for sites which are located at the extreme north and south of the province, ratios of the concentrations between the shutdown period and the operating periods are all less than unity. Some stations (#23 in Figure 5-4 and #23, 37, 21, and 12 in Figure 5-5) along the east side of Georgian Bay have anomalously high ratios and the reason for this is unclear.

5.2.2. N-NO_3^-

Concentration plots for N-NO_3^- are shown in Figures 5-6 to 5-8 for the three periods. Again a negative S to N gradient is observed, but the elevated concentration near Sudbury as observed in the $SO_4^=$ plots is not seen here.

This is consistent with the fact that little NO_x is emitted from the Sudbury sources. The elevated concentration at station 23 (Killarney) is persistent for all three periods but is not easily interpretable.

Figures 5-9 to 5-10 represent ratio plots of N-NO_3^- concentrations for the shutdown vs. the operating periods. It is of interest to note that, except in the vicinity of Sudbury, ratios are in general less than unity on the order of 0.7 to 0.9. The elevation in the Sudbury area is not easily explained, but perhaps can be related to meteorological conditions which are specific to the area.

5.2.3. $SO_4^=$/N-NO_3^-

In view of the fact that both NO_3^- and $SO_4^=$ decrease in concentration to a certain extent during the shutdown period and that the decrease may be attributed to both emissions and meteorological effects, it is desirable to examine ratio plots of $SO_4^=$ to N-NO_3^- (Figures 5-11 to 5-13). One important feature of this approach is that it partially eliminates the effect of meteorology because both $SO_4^=$ and N-NO_3^- experience similar meteorological conditions during the same period. In that sense, N-NO_3^- (which is not emitted from the Sudbury smelters) can be used as a tracer to examine the impact of Sudbury Smelter emissions on $SO_4^=$ concentration. The relatively higher $SO_4^=$/N-NO_3^- ratios around Sudbury with respect to other parts of Ontario during 80/81 and 81/82 but not in 82/83 indicate local impact.

By taking ratios of the ratios of $SO_4^=$/N-NO_3^- for the shutdown period and operating periods, Sudbury emission impact on precipitation quality becomes readily discernible. In Figures 5-14 to 5-15, except in the southern and northern most parts of Ontario, these ratios are less than unity with a minimum (about 0.7) centered around the Sudbury area. The "unity" contour

line should be intrepreted as a boundary beyond which impact due to Sudbury sources in not readily discernible because of variability/uncertainty associated with meteorology and sampling. A further point to note is that the unity contour differs in the two comparison years, suggesting that the results of the Sudbury smelter impact should be regarded as qualitative only.

5.2.4. Cu

Cu, being one of the major emittants characteristic of Cu-Ni smelter operation, is expected to show some definite pattern manifesting smelter impact on precipitation quality. The approach taken here is similar to that taken for $SO_4^=$, i.e. plotting out Cu concentrations for the three periods of interest and ratioing Cu to a tracer ($N\text{-}NO_3^-$).

Figures 5-16 to 5-18 show that a maximum exists around Sudbury with general decreases in locations away from the smelting centre. The mild maximum during the shutdown period is possibly a result of tailings in the surroundings of the smelters. Taking ratios of results of the shutdown period to those of the operating periods leads to contours of less than unity ratio with a minimum around Sudbury, suggesting smelter effects. Plots of Cu-to-NO_3^- ratios which are not included here exhibit a similar pattern to that of the SO_4-to-NO_3^--plots, again pointing to the observed effects of Sudbury smelters.

However, it is somewhat puzzling that ratios in these plots are almost all less than unity across the province and that Ni concentration patterns similar to those of Cu are not observed. Considering that Cu and Ni are both major emittants from the smelters and that they both fall in a similar size range (i.e. similar transport and scavenging patterns are to be expected), the fact that the pattern exists only for Cu and not for Ni, and also the general decrease in the Cu-to-NO_3^- ratio across the province, are observations difficult to reconcile.

5.3. Deposition Contours

Deposition contours of sulfate are given in Figures 5-19 to 5-21 for the three periods and those of their ratios are given in Figrues 5-22 to 5-23. Though the pattern is not as regular as that in the concentration case, nevertheless a decrease in deposition is still observed in the vicinity of Sudbury showing the impact of smelter emissions on wet deposition. Results at station #21 in 1980/81 and at station #23 in 1981/82 and 1982/83 may be anomalous. Similar deposition and deposition ratio plots for N-NO_3^- are shown in Figures 5-24 to 5-26, and 5-27 to 5-28 respectively. There is in general a south to north negative gradient except for stations along the coast of Georgian Bay and part of Lake Huron in the periods of 80/81 and 82/83. The reason for this is unclear but may be related to meteorological factors. Because of this, stations in the neighborhood of Sudbury display larger than unity ratios in the deposition ratio plots between the shutdown year and the preceeding operating years.

5.4. Concluding Remarks

The approach taken here yields qualitative but not quantitative results showing Sudbury emissions impact. This is mainly due to the large variability in the data associated with factors such as meteorology and sampling. Quantitative evaluation of the Sudbury smelter impact on wet deposition can only be obtained by detailed stratification of the event network data, which is the theme of Sections 6 and 7.

6. METEOROLOGICAL ANALYSIS OF DAILY PRECIPITATION DATA

6.1 Dorset Daily Precipitation Data Analysis

Simple trajectory analysis has been carried out using several years of Dorset precipitation data (Kurtz and Yap, 1984). In this section, the contribution of the Sudbury sources is assessed by carrying out a more detailed meteorological stratification. The purpose of this stratification is to minimize the variability associated with other emission sources and meteorological factors, and to isolate the effect of the Sudbury sources (see Appendix 1 for a discussion of the data stratification techniques).

6.1.1 Data Stratification

The Sudbury sources have the highest probability to impact the Dorset site when the synoptic pattern is one of three types: a post-cold front, pre-ridge situation; a cold front passing through the area; or a low pressure system to the northeast (Figure 6-1). Since a cold front is formed due to a cold air mass (flowing toward southeast) pushing and lifting a warm air mass (flowing toward north or northeast), the frontal precipitation samples are affected by sources from south or southeast even when the surface geostrophic wind back trajectories are in the west or northwest sectors. Although the third synoptic type above does not have this problem, the number of available samples is very small. Hence, only the post-cold front, pre-high pressure ridge type is selected here. However, this synoptic pattern may be associated with many different trajectory patterns. In other words, the samples may still be affected by sources from many directions, even for this particular synoptic situation. In order to improve the chance of identifying the Sudbury smelters' effects, we use also trajectory information to stratify the data. By using both the synoptic and trajectory patterns, we are able to minimize both source variability and meteorological variability, and hence, the between-year variation of the stratified data.

6.1.2 Criteria of Selection

According to the above discussion, the data selection criteria are:

(1) post-cold front, pre-high pressure ridge type synoptic pattern;

(2) the envelope of 48 hour back-trajectories calculated every 6 hours during the 24 hour sampling period should include Sudbury;

(3) the 48-hour back-trajectories should at all times be within the 90^{o} sector between west and north (to minimize the possibility of including cases where air parcels with recent history from the high emission density areas to the south have entered the northwest sector).

6.1.3 Data Analysis

6.1.3.1 Selection of the Statistical Test

The significance of the differences of the stratified samples and their concentration means during the shutdown and the operating periods should be examined before using them to estimate the Sudbury smelter shutdown effect. In general, except for testing whether or not there is a difference between populations, all statistical tests for the means are based on the assumption that the samples are random, and most are based on the additional assumption that the populations have normal distributions. Due to the fact that the populations of the present data do not have normal distributions, and each parameter deviates from normality differently, the use of these statistical tests based on a normal distribution will lead to wrong conclusions.

The statistical test of the significance of the means is not necessary if two stratified samples have significantly different population distributions and one sample has a much wider data range than the other, since this condition will provide significantly different means. Since the main purpose of the detailed data stratification is to minimize the meteorological variability and to maximize the Sudbury smelter effect, then for the present analysis to be successful, the

stratified data sets should have distinct data ranges, e.g. the $SO_4^=$ concentration of the shutdown period should be much lower than that of the operating period. In other words, for the successfully stratified data sets, a statistical test of the population differences is sufficient for examining the significance of the Sudbury smelter contribution. For the stratified data sets which do not have distinct data ranges, the evaluation of the Sudbury smelter shutdown effect cannot be performed.

There are many non-parametric tests available in the literature for testing population differences (see Polland, 1977). Since non-parametric tests are robust (not sensitive to slight deviations from the assumed population distribution), all of them should provide similar test results. To justify this point, these methods have been tested by using the $SO_4^=$ and NO_3^- concentrations of the stratified precipitation samples collected in the three periods of interest (July 80 -March 81, July 81 - March 82, and July 82 - March 83) at Dorset. These methods include the Mann-Whitney, Kruskal-Wallis, Median 2-sample, Median one way, Vander Warden, ANOVA and Wilcoxon 2-sample tests. All of these tests provide the same results namely that: (1) the $SO_4^=$ concentrations were significantly lower when the Sudbury smelters were shut down, but the NO_3^- concentrations were not significantly different between shutdown and operating periods; (2) both $SO_4^=$ and NO_3^- are not significantly different between the two operating periods. This result indicates that any of these methods may be used for testing the population differences. In this study, we select the median 2-sample test since it is available in the standard 'SAS' statistical package (SAS, 1979).

6.1.3.2 Significance of the Sudbury Smelter Contribution

According to the criteria of selection described, the number of available cases for the 3 periods of interest (July-March of 80/81, 81/82 and 82/83) is 9, 3 and 5 respectively. (During the second period there were actually 5

meteorologically acceptable occurrences, but on 2 of these occurrences, precipitation chemistry data were missing.) The $SO_4^=$ and $N\text{-}NO_3^-$ concentrations in these samples are plotted in Figure 6-2 (a and b). The significance of the differences among the samples was tested by using the Median 2-sample non-parametric test. The results (see Table 6-1) indicate that the $SO_4^=$ concentrations of the shutdown period are significantly lower than those of the operating periods, while the NO_3^- concentrations of all three periods are not significantly different. Since the Sudbury smelter contribution to NO_3^- is negligible, this observation gives credence to the meteorological stratification scheme. The precipitation depth weighted mean concentrations of these samples are listed in Table 6-2. The precipitation depth-weighted mean $SO_4^=$ concentrations, for the Sudbury smelter operating periods, are comparable, but that for the shutdown period is only about 31 to 33% of those in the operating periods, indicating that the reduction of $SO_4^=$ concentration in the shutdown period is related to the smelter SO_2 emissions.

The Sudbury smelter shutdown effect cannot be shown if one tries to compare directly the deposition data rather than the concentration data. This is due to the variability of the precipitation amount, which introduces a large amount of additional noise into the data analysis. For purposes of illustration, a comparison of deposition similar to that for concentration shown above has been made. The results (Figure 6-3 and Table 6-4) indicate that there is no statistically significant difference of sulfate and nitrate deposition between the smelter shutdown and operating periods (as expected due to the additional "noise" in the depositions data).

6.1.3.3 Estimation of the Sudbury Smelter Contribution to the Wet Deposition at Dorset

The Sudbury contribution to the wet deposition at Dorset for each period of interest was estimated by multiplying the difference of the precipitation depth-

weighted concentrations during the shutdown and operating periods (Table 6-2) with the total precipitation depth accumulated over sampling days during which there were trajectories coming from the Sudbury region (that is, only criterion 2 of Section 6.1.2 had to be satisfied). A number of points should be noted in connection with this calculation:

1. Since many more cases are included in calculating the total precipitation depth associated with the NW quadrant than the limited number of events identified by the rigorous stratification scheme of Section 6.1.2, the total depth used here is considerably greater than the accumulated values of the limited events of Section 6.1.2.

2. The estimate of Sudbury contribution resulting from this calculation is expected to be on the high side, since not every one of the events having a surface geostrophic trajectory in the NW qradrant will be associated with such a high Sudbury impact as in the vigorously stratified data set.

3. The reason that the rigid criteria of Section 6.1.2 were relaxed for this estimate of Sudbury impact on deposition, is the uncertainty about the proper height at which trajectories should be calculated for an elevated source such as the INCO smelter. When the plume stays aloft during the precipitation event, the 850 mb back-trajectory may be more relevant than the surface geostrophic trajectory. In such cases, the 850 mb back-trajectory could be over Sudbury, while the surface geostrophic trajectory is from a more northerly direction. Hence the sector was "widened" to include all the events from the NW quadrant.

Results of this calculation indicate that the Sudbury smelter contribution to the Dorset wet deposition (see Table 6-3) was about 12 and 11% as a probable upper-level estimate for the first and second operating periods, respectively.

6.2 Railton Daily Precipitation Data Analysis

6.2.1 Data Stratification

Due to the fact that the Sudbury smelter contribution to a receptor is inversely proportional to the distance between the receptor and the smelters, it is expected that the Sudbury smelter contribution to Railton is smaller than that at the Dorset site. The chances of identifying the Sudbury smelter effects by meteorological stratification are also smaller. This is due to the fact that there are more important meteorological factors which have to be included in the criteria of data stratification. For study periods of less than a year, as is the case here (since there was only one shutdown period) the chances of success of data stratification are further limited by data availability.

6.2.2 Criteria of Selection

The criteria of data selection for Railton are:

(1) Post-cold front, pre-high pressure ridge type synoptic pattern (See Figure 6-4);

(2) The envelope of 48-hour back trajectories calculated every six hours during the 24-hour sampling period should include Sudbury;

(3) The 48-hour back trajectories should at all times be within the 90° sector between west and north.

The above criteria were used to estimate the contribution of Sudbury smelter to the precipitation concentrations at Railton by comparing values observed during the operating and shutdown periods for the set of samples satisfying these criteria.

As with the Dorset data, an estimate of the contribution of Sudbury to the wet deposition at Railton (probably on the high side) was made by using accumulated depths from all events where the 48-hour back trajectories were, at least one time during the sampling period, within the 90° sector between west and north.

6.2.3 Data Analysis

6.2.3.1 Significance of Sudbury Smelter Contribution

According to the criteria of selection, the number of cases for the three periods of interest (July-March of 80-81, 81-82, and 82-83) are 6, 6, and 3, respectively. It was found that both the $SO_4^=$ and N-NO_3^- concentrations are lower in the shutdown period. Since the Sudbury smelters are not a significant NO_3^- source, the decrease in NO_3^- concentration indicates that factors other than the smelter shutdown are also responsible for the reduction of concentration during the shutdown period. This also suggests that further data stratification is necessary. Unfortunately, this was not possible because the sample size is too small.

The significance of these differences is tested by using the median 2-sample test. Results are listed in Table 6-1. These results suggest that the Sudbury smelter effects are significant for the first operating period, while factors other than emissions from smelters are the main contributors for the second operating period. In other words, the second operating period data should not be used in estimating the Sudbury smelter contribution.

6.2.3.2 Estimation of Sudbury Smelter Contribution to the Wet Deposition at Railton

The Sudbury smelter contribution to the wet deposition at Railton for each period of interest was estimated by using the approach similar to that at Dorset (see section 6.1.3.3). The precipitation depth weighted mean concentration for each period is given in Table 6-2. These results indicate that the precipitation sulfate concentration associated with the most likely Sudbury smelter affected samples is about 12% (from a comparison of the 80-81 and 82-83 data), and the Sudbury smelter contribution is approximately 2% of the total wet deposition for the first operating period (July 1980 - March 1981) (see Table 6-3).

6.3 Longwoods Daily Precipitation Data Analysis

6.3.1 Data Stratification

The synoptic pattern for the cases most likely to be affected by Sudbury is the post cold front pre-high pressure ridge condition when Longwoods is located to the west or northwest of a low pressure system (see Figure 6-5). The 48-hour back trajectories for these cases should be in the north sector (90°).

6.3.2 Criteria of Selection

The criteria of selection for the samples at Longwoods are:

1. post cold front pre-high pressure ridge type synoptic pattern (i.e Longwoods is located to the west or northwest of a low pressure system);
2. the 48-hour back trajectories are in the 90° sector between northwest and northeast.

6.3.3 Data Analysis

6.3.3.1 Significance of Sudbury Smelter Contribution

According to the criteria of selection, the number of cases for the three period of interest (July to March 80-81, 81-82, and 82-83) are 3, 6, and 1, respectively. Since there is only one case in the third period, the significance test was not carried out for the third period. The results (see Table 6-1) of the median 2-sample test indicate that there is no significant difference between the first and the second periods (the operating periods). The estimation of Sudbury smelter contribution to deposition at this site would not be meaningful due to the fact that there is only one case during the shutdown period that falls within the criteria of selection.

6.4 Fernberg Daily Precipitation Data Analysis

Since the Fernberg daily precipitation sampling began only in October 1981, the sample size is not large enough for carrying out the detailed meteorological stratification. Only synoptic patterns were used for data stratification.

6.4.1 Data Stratification and Results

The synoptic patterns corresponding to the most possible Sudbury smelter affected cases are (see Figure 6-6):

1. Fernberg to the northwest of a high pressure system;
2. Fernberg to the southwest of a high pressure system;
3. Fernberg to the northeast of a low pressure system.

There were three cases for the shutdown period, yet only one case for the second operating period and zero for the first period, since no samples were collected. These data do not indicate any shutdown effect since both $SO_4^=$ and N-NO_3^- concentrations are lower in the shutdown period.

6.5 Kapuskasing Daily Precipitation Data Analysis

An attempt was made to examine the smelter impact on precipitation samples collected from the Ontario Hydro's Kapuskasing site. This site is attractive because of its location being north of Sudbury, which is more frequently impacted by air masses passing through Sudbury.

Due to the fact that many precipitation samples of the operating period are missing (from October to December as well as some events in the other months), detailed stratification of the samples was not possible (Jarv, 1982 and Handy, 1984). In total there were only 7 and 16 possible Sudbury smelter-affected cases in the shutdown and operating periods, respectively. However, no similar patterns could be found for these periods. Hence, the analysis was not performed.

6.6 Chalk River Daily Precipitation Data Analysis

6.6.1 Data Stratification

Precipitation data (Barrie et al., 1982) obtained from the Evnironment Canada APN station site at Chalk River, east of Sudbury, were also assessed for Sudbury smelter impact. The synoptic pattern for the most-possible Sudbury smelter-affected cases is given in Figure 6-7. Precipitation samples collected in the shaded area in Figure 6-7 are the ones most probably affected by the Sudbury smelter emissions, since the back-trajectories at Chalk River are then from the Sudbury region.

6.6.2 Criteria of Selection

The criteria of selection of the for Chalk River are:

1. north or northwest of a high pressure system (which is located to the south of a low).
2. the 48-hour back trajectories should be at all times within the west sector (45^o).

These cases were used to estimate the excess Sudbury smelter contribution to the precipitation concentration. The possible Sudbury smelter contribution to the wet deposition at Chalk River was estimated by the product of the excess concentration and the total precipitation depth, using all cases where the 48-hour surface geostrophic wind back trajectories during the sampling period were least one time within the 90^o sector between northwest and southwest.

6.6.3 Data Analysis

6.6.3.1 Significance of the Sudbury Smelter Contribution

In general, the $SO_4^=$ concentrations of the shutdown period are lower than those of the second operating period. However, the NO_3^- concentrations for both periods are in the same range. The results of the median 2-sample test (see

Table 6-1) indicated that the $SO_4^=$ concentrations of the shutdown period are significantly different from those of the second operating period. However, the NO_3^- concentration of the shutdown period are not significantly different from those of the second operating period. There is no significant difference between other periods. These results indicate that the Sudbury smelter contribution to the sulfate precipitation concentration in the second operating period is significant.

6.6.3.2 Estimation of the Sudbury Smelter Contribution to the Wet Deposition at Chalk River

The mean $SO_4^=$ concentration and the percentage difference between the shutdown and operating periods are given in Table 6-3. The Sudbury smelter contribution to the $SO_4^=$ concentration for the cases stratified according to the above criteria is about 49%. There are 49 possible Sudbury smelter-affected cases in the second operating period based on meteorological analysis, however, only 11 samples are available. The total precipitation depth associated with possible effects due to Sudbury is estimated by multiplying the average precipitation depth of the available 11 samples with the total number (49) of possible Sudbury smelter-affected cases. The Sudbury smelter contribution to the wet deposition is then estimated by multiplying this estimated total possible precipitation depth with the excess concentration. Results are given in Table 6-3, indicating that the Sudbury smelter contribution to $SO_4^=$ wet deposition at this site is about 12%.

7. METEOROLOGICAL ANALYSIS OF AIR CONCENTRATION DATA

7.1. Dorset Air Sample Analysis

7.1.1 Data Stratification

Due to the fact that the 6 or 12 hour back-trajectory sector may not be a sufficient indicator of the contributing source and meteorological conditions (see Appendix 1), in this section, the contribution to dry deposition from the Sudbury sources was examined by employing a more detailed data stratification. As discussed in Appendix 1, a specific (48-hour backward) trajectory pattern generally is associated with a specific synoptic pattern variation. Thus the trajectory pattern was here used to take into account both the contributing sources and synoptic pattern variations.

The trajectory pattern for which the Sudbury sources are expected to have the greatest impact at Dorset is the one where trajectories during the sampling period are all bounded by the west and northwest directions (see examples in Figure 7-1). The second most possible trajectory pattern is that which has the 48-hour back-trajectories in the NW sector most of the time during the sampling day (Figure 7-2). The air concentrations are given in Figures 7-3 to 7-5 for events associated with the patterns of this type. From these figures, we can see that during the shutdown period the air $SO_4^=$ and SO_2 concentration were much lower than those of the operating periods. However, the NO_3^- concentrations during the shutdown period were higher than those of the second operating period and slightly lower than those of the first operating period. The median 2-sample test of these samples shows that the shutdown period SO_2 and $SO_4^=$ concentrations are significantly different from those of the operating periods (Table 7-1). However, the NO_3^- concentration of the shutdown period are not significantly different from the second operating period and significantly different from the first operating period. These results indicate that the $SO_4^=$ and SO_2 concentration difference between the shutdown and operating periods may be attributed to a Sudbury

smelter contribution during the second period; however, during the first period differences may have been caused by both Sudbury and other factors. In other words, the Sudbury smelter contribution may be estimated for the second period only. Hence, only the analysis for the second period is given in the next section.

7.1.2. Estimation of the Sudbury Contribution to the Dry Deposition at Dorset.

The contribution of the Sudbury smelters to the dry deposition of SO_2 and $SO_4^=$ at Dorset was estimated by a method similar to that used for wet deposition. Since dry deposition may be estimated by multiplying the appropriate dry deposition velocity with the air concentration, and the dry deposition velocity may be assumed to be constant, on the average, from year to year for the same season, the percentage contribution to dry deposition may be estimated by the percentage contribution to the observed concentration (Table 7-3). By comparing the shutdown (column 7) and operating (column 5) period concentrations for the subset of samples on which the Sudbury sources are expected to have the greatest impact, the excess concentration due to the Sudbury source was obtained in column 8. This concentration was then multiplied with the total number of cases where Sudbury could have had a possible impact on Dorset (column 10) and divided by the average concentration measured during the operating period times the total number of measurements during the operating period. The criterion of selecting all the possible Sudbury affected cases is that the 48-hour back trajectories should at least one time during the sampling period be within the 90^o sector between west and north. This criterion takes into account upper level trajectories including all the possible Sudbury effects on air concentration through direct transport, and indirect mixing from the surface to the 850 mb level. The ratio, expressed as a percentage, is shown in the last column, and is an estimate (probably on the high side) of the Sudbury source contribution to the dry deposition at Dorset.

Results indicate that the Sudbury sources contributed about 88% of the SO_2 concentration and 81% of the $SO_4^=$ concentration of the stratified samples during the second operating period. An upper estimate of the Sudbury source contribution to the dry SO_2 deposition at Dorset site is about 31% for the period from July 1981 to March 1982. The Sudbury source contribution to the dry $SO_4^=$ deposition is about 18% for the second period of interest.

7.2 Railton Air Sample Analysis

7.2.1 Data Stratification

The synoptic patterns for the most probable Sudbury smelter-affected cases at Railton are similar to those for the Dorset data. These trajectory patterns are given in Figure 7-2.

7.2.2 Data Analysis

7.2.2.1 Significance of the Sudbury Smelter Contribution

Since the sample size of the first period (80/81) is too small for the detailed data stratification, the estimation of Sudbury smelter contribution for this period is not given here.

Results of the median 2-sample test (see Table 7-1) indicate that the SO_2 concentrations of the shutdown period (82/83) are significantly lower than those of the second operating period (81/82). However, in the cases of $SO_4^=$ and N-NO_3^-, they are not significantly different. In other words, the Sudbury smelters contribute a significant amount of SO_2 to Railton, but their contribution of $SO_4^=$ is relatively small.

7.2.2.2 Estimation of the Sudbury Smelter Contribution to the Dry Deposition at Railton

The mean SO_2 concentration and the percentage difference between the shutdown and operating periods are given in Table 7-3. The Sudbury smelter

contribution to the air SO_2 concentration at Railton is about 86%. The Sudbury smelter contribution to the dry deposition is estimated by using the approach similar to that in section 7.1.2. The estimated Sudbury smelter contribution to the SO_2 dry deposition at Railton is about 27%. Again, this estimate is thought to be on the high side, for reasons already discussed in connection with the Dorset analysis.

7.3. Longwoods Air Sample Analysis

7.3.1 Data Stratification

The synoptic patterns for the most possible Sudbury smelter-affected cases at Longwoods are given in Figure 6-5. The criteria of selection of the most possible Sudbury smelter affected cases are:

(1) north or northwest of a low type synoptic pattern;

(2) the 48-hour back-trajectory should at all time be within the north sector (45^o).

The criterion used for selecting the possible Sudbury smelter affected cases is that the 48-hour back trajectories should at least one time during the sampling period be in the north sector (45^o sector).

7.3.2 Data Analysis

7.3.2.1 Significance of the Sudbury Smelter Contribution

Since the sample size of the first period (80/81) is not large enough, the analysis is not given here.

Results of the median 2-sample test (see Table 7-1) indicate that only SO_2 concentrations but not $SO_4^=$ and NO_3^- are significantly different between the Sudbury smelter operating (81/82) and shutdown (82/83) periods.

7.3.2.2 Estimation of the Sudbury Smelter Contribution to the Dry Deposition at Longwoods

The mean air SO_2 concentration and the percentage difference between the shutdown and operating periods are given in Table 7-3. The Sudbury smelter contribution to the air SO_2 concentration at Longwoods is about 74% (for the stratified data set). The estimated Sudbury smelter contribution to the SO_2 dry deposition at this site is about 5% (see Table 7-3), a value probably on the high side as discussed above.

7.4 Fernberg Air Sample Analysis

7.4.1 Data Stratification

Due to the fact that the sample size is too small after being stratified by both the trajectory pattern and the synoptic pattern, only the synoptic pattern is used to select the most possible Sudbury smelter affected cases. The criteria of selection of the most possible Sudbury affected cases are: that Fernberg should be to the northwest of a high pressure system and that the envelope of the 48-hour back-trajectories calculated every 6 hours during the 24-hour sampling period should include Sudbury.

7.4.2 Data Analysis

7.4.2.1 Significance of the Sudbury Smelter Contribution

Due to the limited sample size of the first operating period (80/81), the analysis for this period is not given here.

The results of the median 2-sample test of the significance of the difference between the samples collected during the operating (81/82) and the shutdown (82/83) periods are given in Table 7-1. These results indicate that only the air SO_2 concentrations are significantly different between the two periods. If we assume that this difference is mainly due to the Sudbury smelter shutdown (we

do not have enough meteorological information to justify this), we may then estimate the Sudbury smelters contribution to air concentration for the stratified data, and to dry deposition at Fernberg. This estimation is given in the next section.

7.4.2.2. Estimation of the Sudbury Smelter Contribution to Dry Deposition at Fernberg

The result of the calculations is given in Table 7-3. This result indicates that Sudbury smelter contributed to the air SO_2 concentration at Fernberg is about 57%. By using the approach similar to that for the other sites we may estimate the possible Sudbury smelter contribution to the total dry deposition. Since samples are available only after October, 1981, an estimate is made for the period from October 1, 1981 to March 31, 1982 (see Table 7-2). These results indicate that the Sudbury smelters contribute about 4% of the total SO_2 dry deposition (from October 1, 1981 to March 30, 1982) at Fernberg.

7.5 Kapuskasing Air Sample Analysis

7.5.1 Data Stratification

The synoptic patterns for the possible Sudbury smelter-affected cases at Kapuskasing (Ontario Hydro network site) are west or northwest of a high pressure system, post-ridge/pre-warm front; warm sector (with southerly flows); and pre-north-south oriented cold front (see Figure 7-6). Of these, the case where Kapuskasing is west or northwest of a high pressure system is the one which occurs most frequently.

7.5.2 Data Analysis

7.5.2.1 Significance of the Sudbury Smelter Contribution

Since only the second and third periods have enough data for analysis, the following analysis is for those two periods only.

The NO_3^- concentration of the stratified samples for both the operating and shutdown periods are below the analytical detection limit, and no meaningful comparison can be made. Results of the median 2-sample test (see Table 7-1) indicate that the SO_2 and $SO_4^=$ concentrations during the shutdown period are significantly different from those of the 81/82 operating period. These results indicate that the Sudbury smelter contribution is significant.

7.5.2.2 Estimation of the Sudbury Smelter Contribution to the Dry Deposition at Kapuskasing

The mean SO_2 and $SO_4^=$ concentration and the percentage difference between the shutdown and operating periods are given in Table 7-3. The Sudbury smelter contribution to the SO_2 and $SO_4^=$ concentrations at Kapuskasing is about 92 and 66% respectively. The estimated Sudbury smelter contribution to dry deposition at Kapuskasing is given in Table 7-3, namely, about 47% and 46%, respectively for SO_2 and $SO_4^=$, which values are again expected to be near upper limits, due to the nature of the present analysis.

7.6 Chalk River Daily Air Sample Analysis

7.6.1 Data Stratification

The synoptic pattern and trajectory patterns of the most possible Sudbury smelters affected cases given in Figure 6-7. In these cases, the pollutants emitted from the Sudbury smelters are transported to the Chalk River (Environment Canada APN site) area.

7.6.2 Criteria of Selection

The criteria of selection of the most possible Sudbury smelters affected cases are:

1. north or northwest of a high pressure system;

2. the 48-hour back trajectories should at all times be within the west sector (45^{o}).

To calculate an upper estimate of the contribution of Sudbury smelters to dry deposition, cases were used where the 48-hour surface geostrophic back trajectories during the 24-hour sampling period were at least one time within the west sector between northwest and southwest. Since in general, a westerly 850 mb-level back trajectory may be associated with a northwesterly to westerly surface geostrophic wind trajectory when a cold front is passing through and with a southwesterly to westerly surface geostrophic trajectory when a warm front is moving away, this criterion should be sufficient for considering the possibility of Sudbury smelter contribution from surface to 850 mb-level.

7.6.3 Data Analysis

7.6.3.1 Significance of the Sudbury Smelter Contribution

In general, the air concentrations of the shutdown period are lower than those of the operating period. Results of the median 2-sample test (Table 7-1) indicate that the SO_2 and $SO_4^=$ concentrations of the shutdown period are significantly lower than those of the operating periods. However, the NO_3^- concentration of the shutdown period is not significantly different from that of the operating period. These results indicate that the Sudbury smelter shutdown is the major factor responsible for concentration reduction during the shutdown period.

7.6.3.2 Estimation of the Sudbury Smelter Contribution to the Dry Deposition at Chalk River

The mean SO_2 and $SO_4^=$ concentrations and the percentage difference between the shutdown and operating period are given in Table 7-3. The Sudbury smelter contributions to the SO_2 and $SO_4^=$ concentration (for the most probable Sudbury smelter-affected cases) at Chalk River for the first operating period are

about 87% and 55%, and for the second period are about 85% and 47% respectively. The estimation of the Sudbury smelter contribution to the dry deposition at Chalk River is given in Table 7-3. Results indicate that the Sudbury smelter contributions to the dry SO_2 and $SO_4^=$ deposition for the first operating period are about 25% and 17% and for the second period are about 26% and 20% respectively.

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S T A T I S T I C A L   A N A L Y S I S   S Y S T E M

TABLE 4-1 SUMMARY OF DATA USED IN ANALYSIS

		# of data points used					H_f^+	H_t^+	$SO_4^=$	$N-NO_3^-$	Cu	# of missing data points				
OBS	Period	N1*	N2	N3	N4	N5	MEAN1	MEAN2	MEAN3	MEAN4	MEAN5	MISS1*	MISS2	MISS3	MISS4	MISS5
1	8008	18	18	17	17	8	0.0704280	0.094068	3.32747	0.353679	0.0033215	0	0	1	1	10
~~2~~	~~8011~~	~~22~~	~~20~~	~~23~~	~~23~~	~~17~~	~~0.0373767~~	~~0.074960~~	~~2.08627~~	~~0.433857~~	~~0.0027564~~	~~4~~	~~6~~	~~3~~	~~3~~	~~9~~
3	8102	24	17	24	22	16	0.0522937	0.082553	2.47350	0.516367	0.0036494	3	10	3	5	11
4	8105	24	26	24	27	19	0.0472944	0.082538	3.93586	0.477630	0.0040550	1	1	3	0	8
5	8108	27	27	26	26	22	0.0747589	0.107562	3.86520	0.380928	0.0023669	1	1	2	2	6
6	8111	27	27	27	27	24	0.0427766	0.079035	1.66727	0.360606	0.0042136	3	3	3	3	6
7	8202	27	22	27	27	18	0.0577584	0.089163	1.80737	0.552883	0.0139957	2	7	2	2	11
~~8~~	~~8205~~	~~30~~	~~29~~	~~28~~	~~29~~	~~26~~	~~0.0848493~~	~~0.100071~~	~~4.18406~~	~~0.583528~~	~~0.0017553~~	~~0~~	~~1~~	~~2~~	~~1~~	~~4~~
9	8208	39	38	38	40	38	0.0667452	0.084961	2.89455	0.379009	0.0010934	2	3	3	1	3
10	8211	27	27	27	25	28	0.0447867	0.068307	1.89182	0.352788	0.0023004	3	3	3	5	2
11	8302	25	26	27	26	27	0.0615024	0.087506	2.18864	0.512880	0.0017886	5	4	3	4	3

*Parameters: 1=H_f^+, 2=H_t^+, 3=$SO_4^=$, 4=$N-NO_3^-$ and 5= Cu. (in mg/l unit)

Table 4-2

Statistical Significance at 95% Confidence Level Using Median 2-Sample Test*

	July - Sept			Oct. - Dec.			Jan. - Mar.		
	1980 vs. 81	1980 vs 82	1981 vs 82	1980 vs 81	1980 vs 82	1981 vs 82	1981 vs 82	1981 vs 83	1982 vs 83
H_f^+	No (.054)	No (.544)	Yes (.041)	No (.160)	No (.321)	No (.590)	No (.493)	No (.160)	No (.590)
H_t^+	No (.092)	No (.460)	Yes (.001)	No (.388)	No (.422)	Yes (.015)	No (.414)	Yes (.010)	No (1.0)
$SO_4^=$	No (.156)	No (.756)	Yes (.018)	No (.072)	NO (.214)	No (.590)	No (.072)	No (.327)	No (.281)
$N\text{-}NO_3^-$	No (.852)	No (.760)	No (.218)	No (.695)	No (1.00)	No (.267)	No (.898)	No (.119)	No (.894)
Cu	No (.104)	No (1.00)	Yes (.022)	No (.855)	Yes (.001)	Yes (.000)	Yes (.007)	Yes (.001)	No (.054)

* Values in brackets represent probability that the comparison periods' results are not different.

TABLE 5.1

APIOS Cumulative Network Station Identification

STATIONS

1	COLCHESTER	23	KILLARNEY
2	MERLIN	24	BEAR ISLAND
3	PORT STANLEY	25	GOWGANDA
4	WILKESPORT	26	RAMSEY
5	ALVINSTON	27	MOONBEAM
6	HURON PARK	28	ATTAWAPISKAT
7	WATERLOO	29	WINISK
8	PALMERSTON	30	NAKINA
9	SHALLOW LAKE	31	DORION
10	MILTON	32	QUETICO CENTRE
11	UXBRIDGE	33	LAC LA CROIX
12	COLDWATER	34	E.L.A.
13	CAMPBELLFORD	35	EAR FALLS
14	KALADAR	36	PICKLE LAKE
15	SMITH'S FALLS	37	BURWASH
16	DALHOUSIE MILLS	38	LIVELY
17	GOLDEN LAKE	39	HANMER
18	WILBERFORCE		
19	WHITNEY		
20	DORSET		
21	MCKELLAR		
22	MATTAWA		

Table 6-1

Results of the Median 2-Sample Test of the Significance of the Difference (precipitation concentration) Between the Most Possible Sudbury Affected Samples Collected During the Sudbury Shutdown and Operating Periods*

Site	Comparison Periods	Parameter	Test Value Z	Probability >|Z|	Significant
Dorset	80/81 vs 82/83	$SO_4^=$	-2.13	0.03	Yes
		$N\text{-}NO_3^-$	-0.98	0.33	No
	81/82 vs 82/83	$SO_4^=$	1.82	0.07	Yes
		$N\text{-}NO_3^-$	1.01	0.31	No
	80/81 vs 81/82	$SO_4^=$	-0.64	0.52	No
		$N\text{-}NO_3^-$	0.64	0.52	No
Railton	80/81 vs 82/83	$SO_4^=$	-1.78	0.08	Yes
		$N\text{-}NO_3^-$	-0.44	0.65	No
	81/82 vs 82/83	$SO_4^=$	-0.44	0.65	No
		$N\text{-}NO_3^-$	-1.78	0.08	Yes
	80/81 vs 81/82	$SO_4^=$	1.11	0.27	No
		$N\text{-}NO_3^-$	1.11	0.27	No
Longwoods	80/81 vs 81/82	$SO_4^=$	0.89	0.37	No
		$N\text{-}NO_3^-$	0.89	0.37	No
Chalk River	80/81 vs 82/83	$SO_4^=$	1.49	0.14	No
		$N\text{-}NO_3^-$	0.75	0.46	No
	81/82 vs 82/83	$SO_4^=$	-2.05	0.04	Yes
		$N\text{-}NO_3^-$	-0.68	0.49	No
	80/81 vs 81/82	$SO_4^=$	-0.68	0.49	No
		$N\text{-}NO_3^-$	-0.68	0.49	No

* Shutdown period: July 1982 - March 1983. Operating Period: July 1980 - March 1981 and July 1981 - March 1982.

Table 6.2

Precipitation-depth-weighted Mean and Standard Deviation
of $SO_4^=$ and $N\text{-}NO_3^-$ Concentration (mg/l) of the
Most Possible Sudbury Smelter Affected Cases

Site	Period*	N	Precipitation Total Depth (mm)	$SO_4^=$ Mean	$SO_4^=$ S.D.	$N\text{-}NO_3^-$ Mean	$N\text{-}NO_3^-$ S.D.
	80 - 81	9	20.1	1.36	0.64	0.24	0.15
Dorset	81 - 82	3	11.6	1.46	0.41	0.18	0.04
	82 - 83	4	11.4	0.45	0.34	0.14	0.16
	80 - 81	6	9.7	1.92	0.93	0.84	0.35
Railton	81 - 82	6	16.8	2.31	0.86	0.89	0.38
	82 - 83	3	19.6	1.69	0.84	0.40	0.19
	80 - 81	3	1.35	2.50	0.38	0.42	0.13
Longwoods	81 - 82	6	39.7	2.49	0.28	0.27	0.10
	82 - 83	1	9.9	1.60	-	0.20	-
	80 - 81	3	13.2	6.60	3.70	3.03	2.01
Chalk River	81 - 82	5	49.5	3.36	1.49	1.67	0.82
	82 - 83	3	17.7	1.73	0.43	1.13	0.57

* All periods refer to July of the first year to March of the second year.

Table 6.3

Estimated Sudbury Smelter Contribution to Precipitation Concentration and Wet Deposition

			(1)		(2)	(3)=(1)-(2)	100x(3)/(1)		(4)	(5)=(3)x(4)	(6)	(7)=100x(5)/(6)
Site	Period*	MPSAC+ Average $SO_4^=$ concentration (mg l^{-1})				Excess $SO_4^=$ (mg l^{-1})	Sudbury contribution to $SO_4^=$ concentration (%)	Possible Sudbury affected cases		Sudbury contribution $SO_4^=$ Wet deposition (mg m^{-2})	Total period Wet $SO_4^=$ deposition (mg m^{-2})	Sudbury smelters' contribution $SO_4^=$ wet deposition (%)
		N	Operating	N	Shutdown			N	Total precipitation (mm)			
Dorset	80/81 vs 82/83	9	1.36	4	0.45	0.91	67	52	291	264.8	2202.7	12
	81/82 vs 82/83	3	1.46	4	0.45	1.01	69	52	250	252.5	2229.3	11
Railton	80/81 vs 82/83	6	1.92	3	1.69	0.23	12	38	187.4	43.1	2050.0	2
Chalk River	81/82 vs 82/83	5	3.36	3	1.73	1.63	49	49	79.8	227.0	1955.0	12

* All periods refer to July of the first year to March of the second year as indicated.

\+ MPSAC = Most Possible Sudbury Smelters Affected Cases.

Table 6-4

Results of Median 2-Sample Test of the
Significance of the Difference (Deposition) at Dorset
Between the Sudbury Smelter Shutdown and Operating Periods*

Test Period	Test Parameter	Test Value Z	Probability >|Z|	Significant
(80-81 and 81-82) vs (82-83)	SO_4	-0.365	0.176	No
(80-81) vs (81-82)	SO_4	-0.638	0.523	No
(80-81 and 81-82) vs (82-83)	$N\text{-}NO_3$	-0.365	0.715	No
(80-81) vs (81-82)	$N\text{-}NO_3$	0.638	0.523	No

* All periods refer to July of the first year to March of the second year as indicated.

Table 7.1

Results of the Median 2-Sample Test of the Significance of the Difference of the Air Concentration Between the Shutdown and Operating periods

Site	Period*	Parameter	Test Value Z	Probability $>\lvert Z\rvert$	Significant
Dorset	80/81 vs 82/83	$SO_4^=$	-2.77	0.01	Yes
		$N\text{-}NO_3^-$	-1.84	0.07	Yes
	81/82 vs 82/83	$SO_4^=$	-2.77	0.01	Yes
		$N\text{-}NO_3^-$	1.16	0.24	No
		SO_2	-1.66	0.10	Yes
	80/81 vs 81/82	$SO_4^=$	0	1.0	No
		$N\text{-}NO_3^-$	-1.55	0.12	No
Railton	81/82 vs 82/83	$SO_4^=$	-1.08	0.28	No
		$N\text{-}NO_3^-$	-1.24	0.22	No
		SO_2	-2.54	0.01	Yes
Longwoods	81/82 vs 82/83	$SO_4^=$	-1.41	0.16	No
		$N\text{-}NO_3^-$	-1.41	0.16	No
		SO_2	-3.15	0.002	Yes
Fernberg	81/82 vs 82/83	$SO_4^=$	-0.40	0.68	No
		$N\text{-}NO_3^-$	1.01	0.31	No
		SO_2	2.42	0.02	Yes
Kapuskasing	81/82 vs 82/83	$SO_4^=$	2.34	0.025	Yes
		SO_2	2.34	0.025	Yes
Chalk River	80/81 vs 82/83	$SO_4^=$	-2.45	0.01	Yes
		$N\text{-}NO_3^-$	-0.98	0.33	No
		SO_2	-2.45	0.01	Yes
	81/82 vs 82/83	$SO_4^=$	-2.25	0.02	Yes
		$N\text{-}NO_3^-$	-0.98	0.33	No
		SO_2	-2.25	0.02	Yes
	80/81 vs 81/82	$SO_4^=$	-0.17	0.14	No
		$N\text{-}NO_3^-$	-0.60	0.55	No
		SO_2	0.84	0.40	No

* All periods refer to July of the first year and March of the second year as indicated.

Table 7.2

Mean and Standard Deviation of the Air Concentration (ug/m^3) of the Most Possible Sudbury Smelter Affected Cases

Site	Period*		$SO_4^=$			NO_3^-			SO_2	
		N	Mean	S.D.	N	Mean	S.D.	N	Mean	S.D.
Dorset	80 - 81	10	1.67	1.0	10	0.012	0.006	1	11.23	-
	81 - 82	10	1.73	1.1	9	0.016	0.018	9	4.53	4.07
	82 - 83	8	0.33	0.28	8	0.013	0.006	8	0.53	0.51
Railton	81 - 82	16	1.71	1.47	16	0.08	0.10	16	4.66	5.74
	82 - 83	9	1.09	0.96	10	0.14	0.19	9	15.3	13.5
Longwoods	81 - 82	11	2.31	1.46	11	0.20	0.15	10	3.47	3.35
	82 - 83	7	1.08	0.56	7	0.10	0.08	7	0.90	0.51
Fernberg	81 - 82	3	0.74	0.39	3	0.007	0.006	3	2.65	1.13
	82 - 83	4	1.26	0.52	4	0.003	0.005	4	1.15	0.19
Kapuskasing	81 - 82	3	6.15	3.60	3	0.80	0.57	3	8.17	5.40
	82 - 83	3	2.08	0.55	3	0.1	0.06	3	0.67	0.12
Chalk River	80 - 81	6	1.62	0.44	5	0.61	0.35	6	6.15	5.12
	81 - 82	5	1.38	0.81	5	0.50	0.36	5	5.36	2.89
	82 - 83	4	0.73	0.10	4	0.27	0.13	4	0.83	0.59

* All periods refer to July of the first year and March of the second year as indicated.

Table 7.3

Estimated Sudbury Smelter Contribution to Dry Deposition and Air Concentration

Site	Period+	Parameter	(1) N	(1) MPSAC* Average Operating	(2) N	(2) Concentration Shutdown	(3)=(1)-(2) Excess (ug/m^3)	100x(3)/(1) Sudbury contribution to air concentration	(4) N Possible Sudbury affected cases	(5)=(3)x(4) Estimated Sudbury contrib.(ug/m^3)	(6) Total concentration of the period (ug/m^3)	(7)=100x(5)/(6) Sudbury Smelter contribution (%)
Dorset	80 - 81	SO_2	(4)	-	(10)	-	4.0**	--	170	680	1842	37
		$SO_4^=$	(10)	1.67	(8)	0.33	1.34	80	170	228	810	28
	81 - 82	SO_2	(9)	4.53	(10)	0.53	4.0	88	127	508	1616	31
		$SO_4^=$	(10)	1.73	(8)	0.33	1.40	81	127	177	969	18
Railton	81 - 82	SO_2	(16)	4.66	(8)	0.66	4.0	86	127	508	1902	27
Longwoods	81 - 82	SO_2	(10)	3.47	(7)	0.90	2.57	74	68	175	3250	5
Fernberg	81 - 82	SO_2	(3)	2.65	(4)	1.15	1.5	57	13	19.5	492	4
Kapuskasing	81 - 82	SO_2	(3)	8.17	(3)	0.67	7.5	92	46	345	737	47
		$SO_4^=$	(3)	6.15	(3)	2.08	4.07	66	46	187	406	46
Chalk River	80 - 81	SO_2	(6)	6.15	(4)	0.83	5.32	87	121	644	2572	25
		$SO_4^=$	(6)	1.62	(4)	0.73	0.89	55	121	108	625	17
	81 - 82	SO_2	(4)	5.36	(4)	0.83	4.53	85	142	643	2468	26
		$SO_4^=$	(4)	1.38	(4)	0.73	0.65	47	142	92.3	468	20

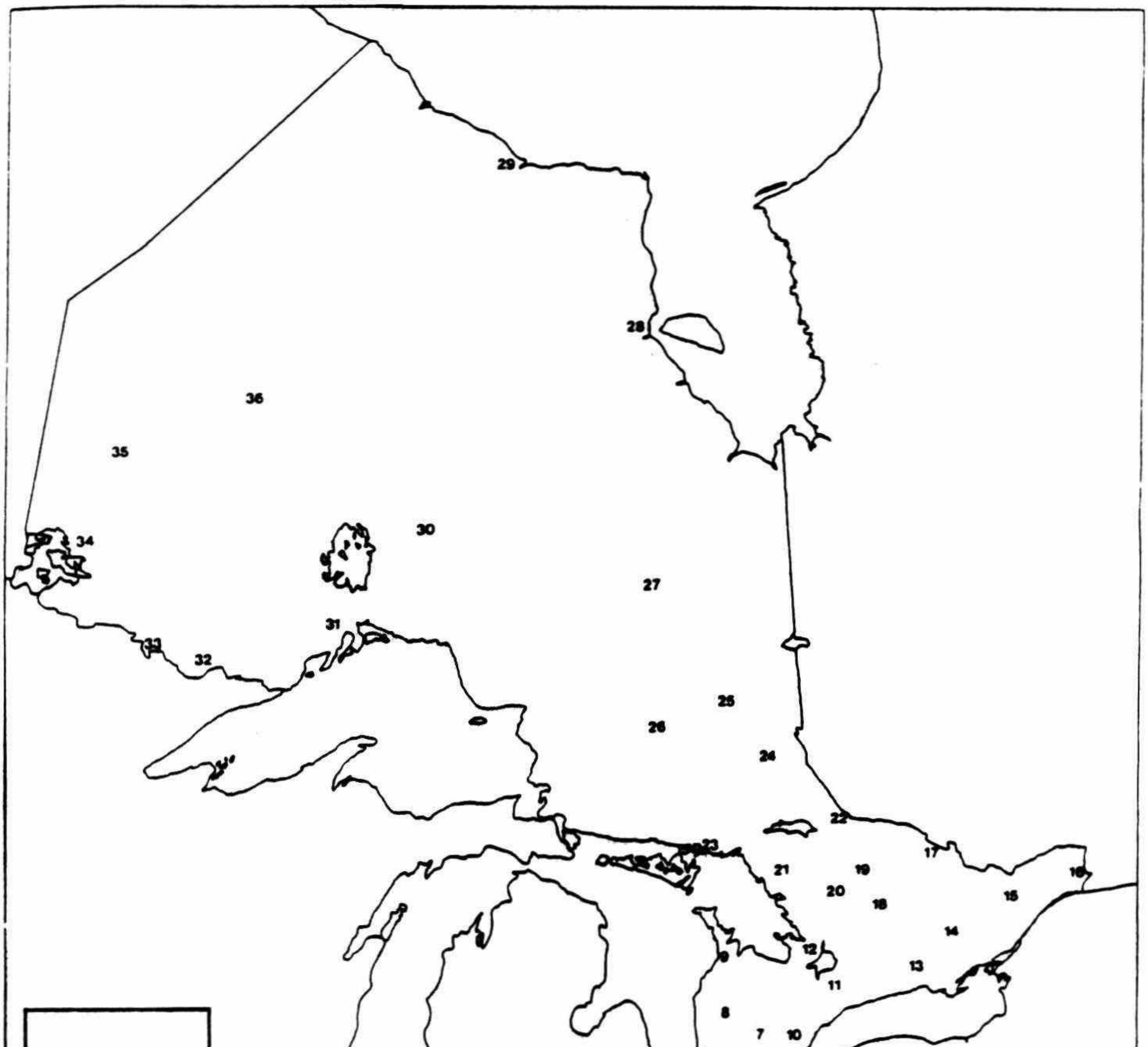
* MPSAC = most possible Sudbury affected case

\+ All periods refer to July of the first year to March of the second year as indicated.

** The 80/81 excess concentration is assumed to be the same as that of 81/82.

FIGURE 2 - 1 : STATION LOCATION MAP

CUMULATIVE PRECIPITATION
MONITORING NETWORK

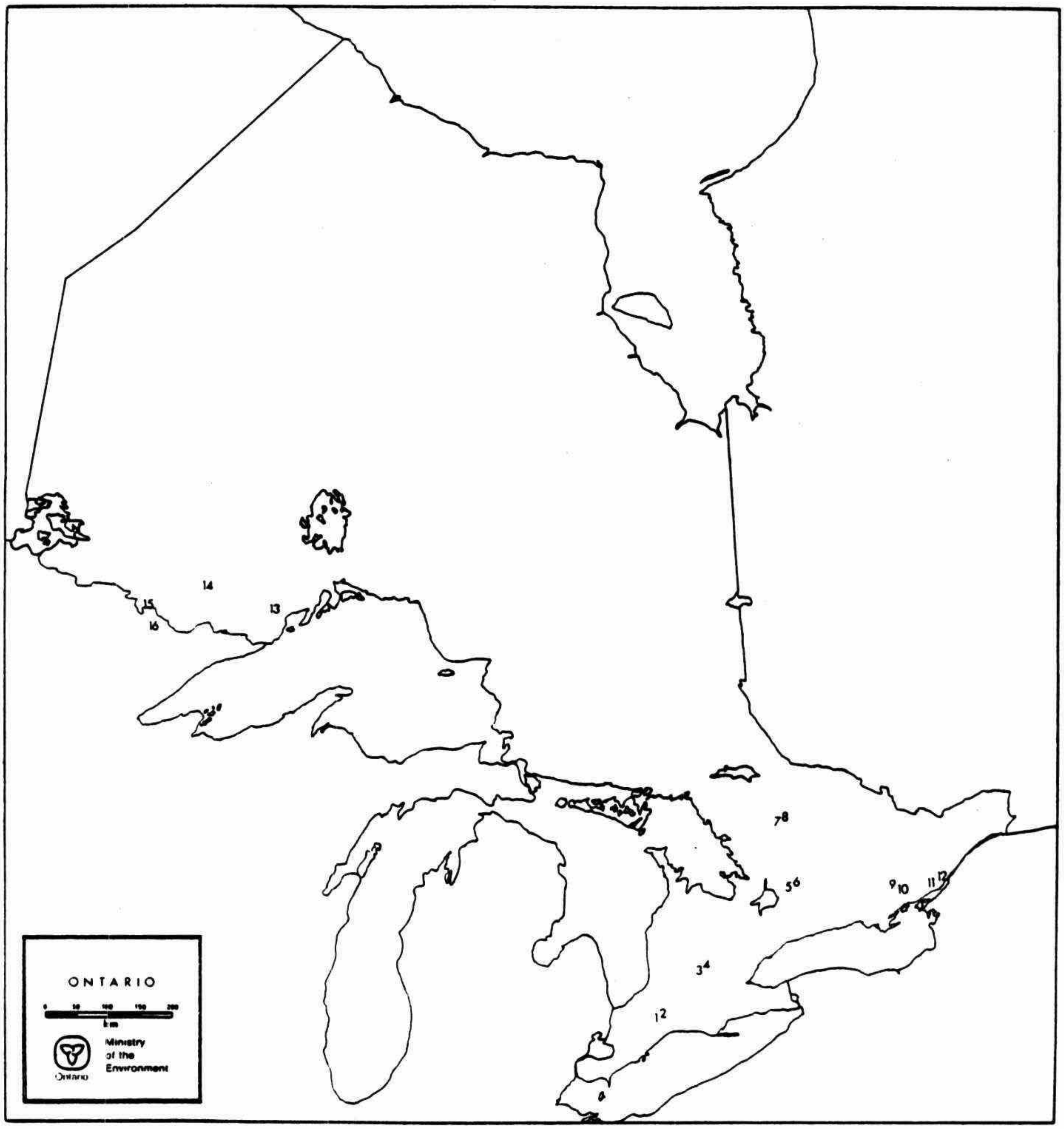


1 - COLCHESTER
2 - MERLIN
3 - PORT STANLEY
4 - WILKESPORT
5 - ALVINSTON
6 - HURON PARK
7 - WATERLOO
8 - PALMERSTON
9 - SHALLOW LAKE
10 - MILTON
11 - UXBRIDGE
12 - COLDWATER
13 - CAMPBELLFORD
14 - KALADAR
15 - SMITH'S FALLS
16 - DALHOUSIE MILLS
17 - GOLDEN LAKE
18 - WILBERFORCE
19 - WHITNEY
20 - DORSET
21 - MCKELLAR
22 - MATTAWA
23 - KILLARNEY
24 - BEAR ISLAND
25 - GOWGANDA
26 - RAMSEY
27 - MOONBEAM
28 - ATTAWAPISKAT
29 - WINISK
30 - NAKINA
31 - DORION
32 - QUETICO CENTRE
33 - LAC LA CROIX
34 - EXP. LAKES AREA
35 - EAR FALLS
36 - PICKLE LAKE

FIGURE 2 - 2

STATION LOCATION MAP

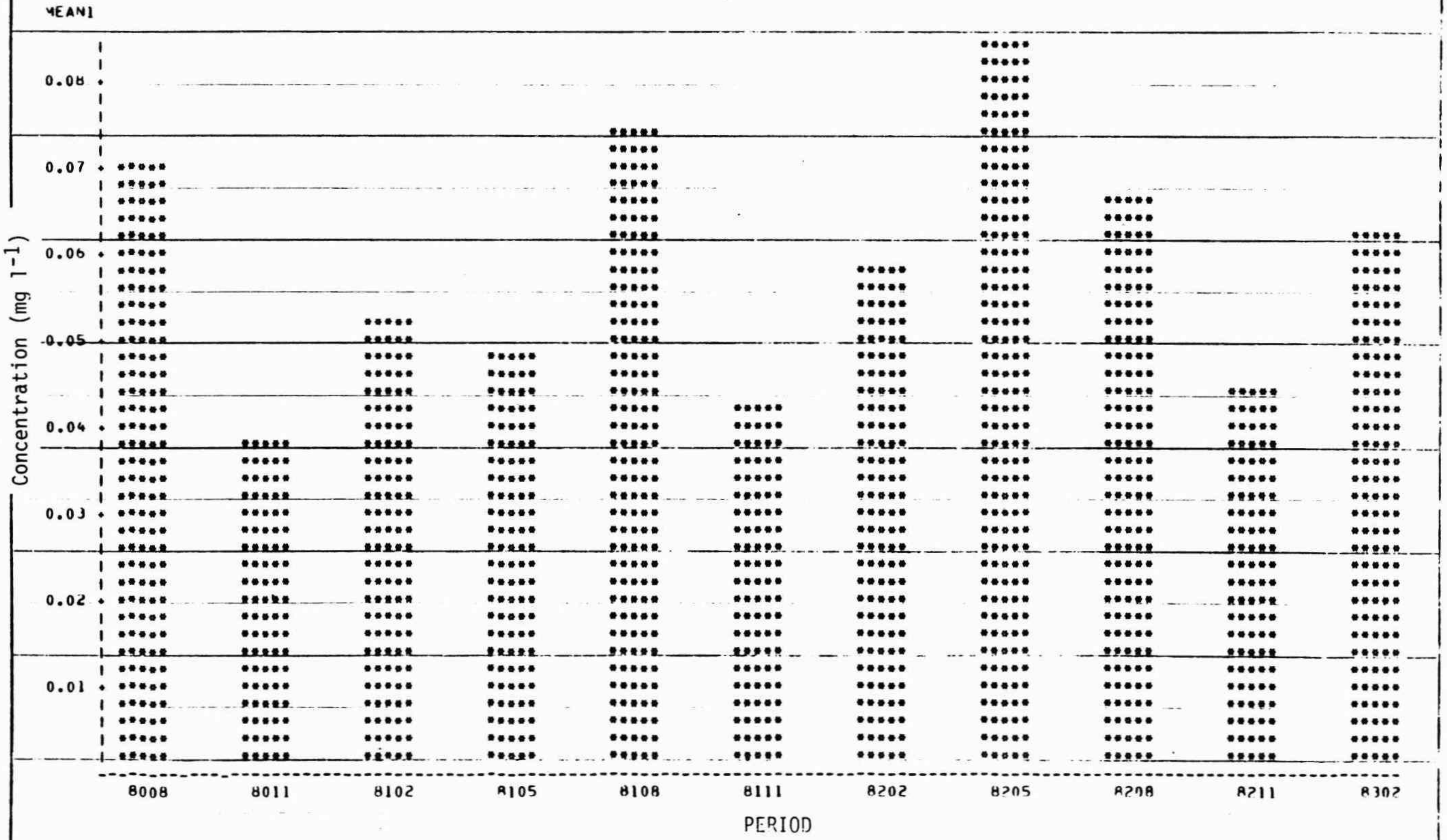
DAILY PRECIPITATION/AIR* MONITORING NETWORK



1 - MELBOURNE	5 - RAVEN LAKE	9 - WHITMAN CREEK	13 - FORBES TOWNSHIP
* 2 - LONGWOODS	6 - BALSAM LAKE	10 - RAILTON	14 - QUETICO CENTRE
3 - NORTH EASTHOPE	7 - NITHGROVE	* 11 - CHARLESTON LAKE	15 - LAC LA CROIX
4 - WELLESLEY	* 8 - DORSET	12 - GRAHAM LAKE	* 16 - FERNBERG

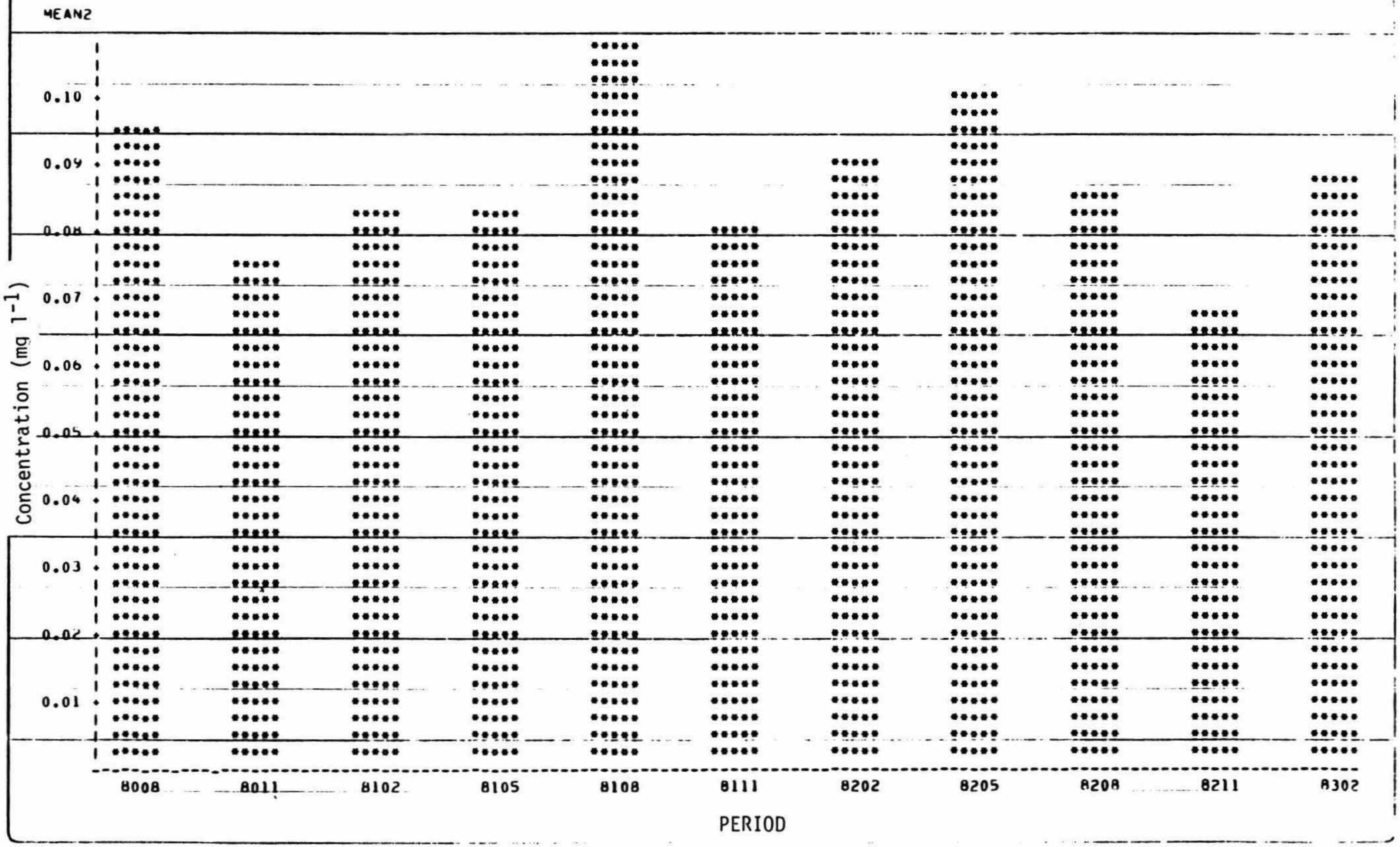
STATISTICAL ANALYSIS SYSTEM

FIGURE 4-1 : BAR CHART OF MEAN1 (H_f^+)



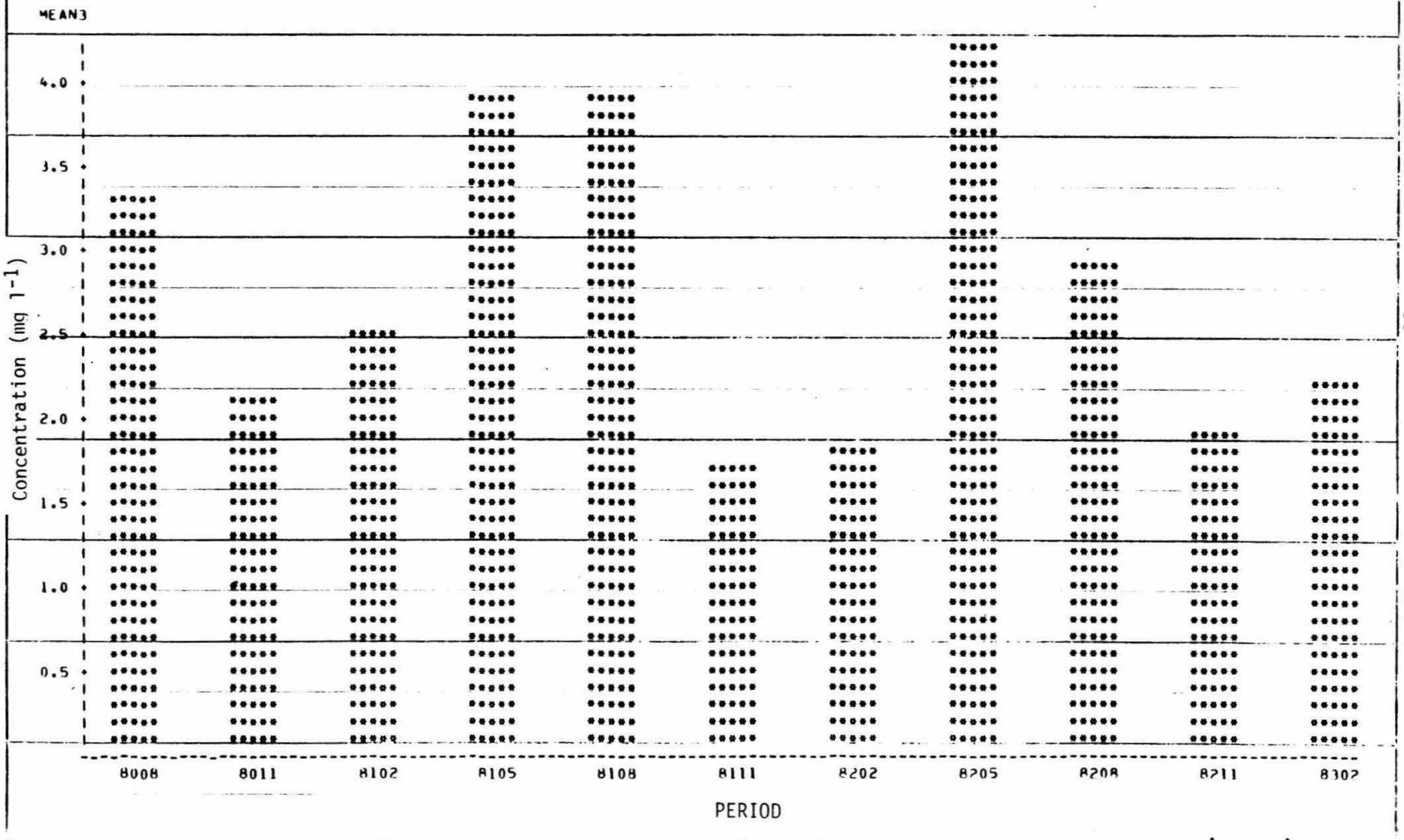
STATISTICAL ANALYSIS SYSTEM

FIGURE 4-2 BAR CHART OF MEAN2 (H_t^+)



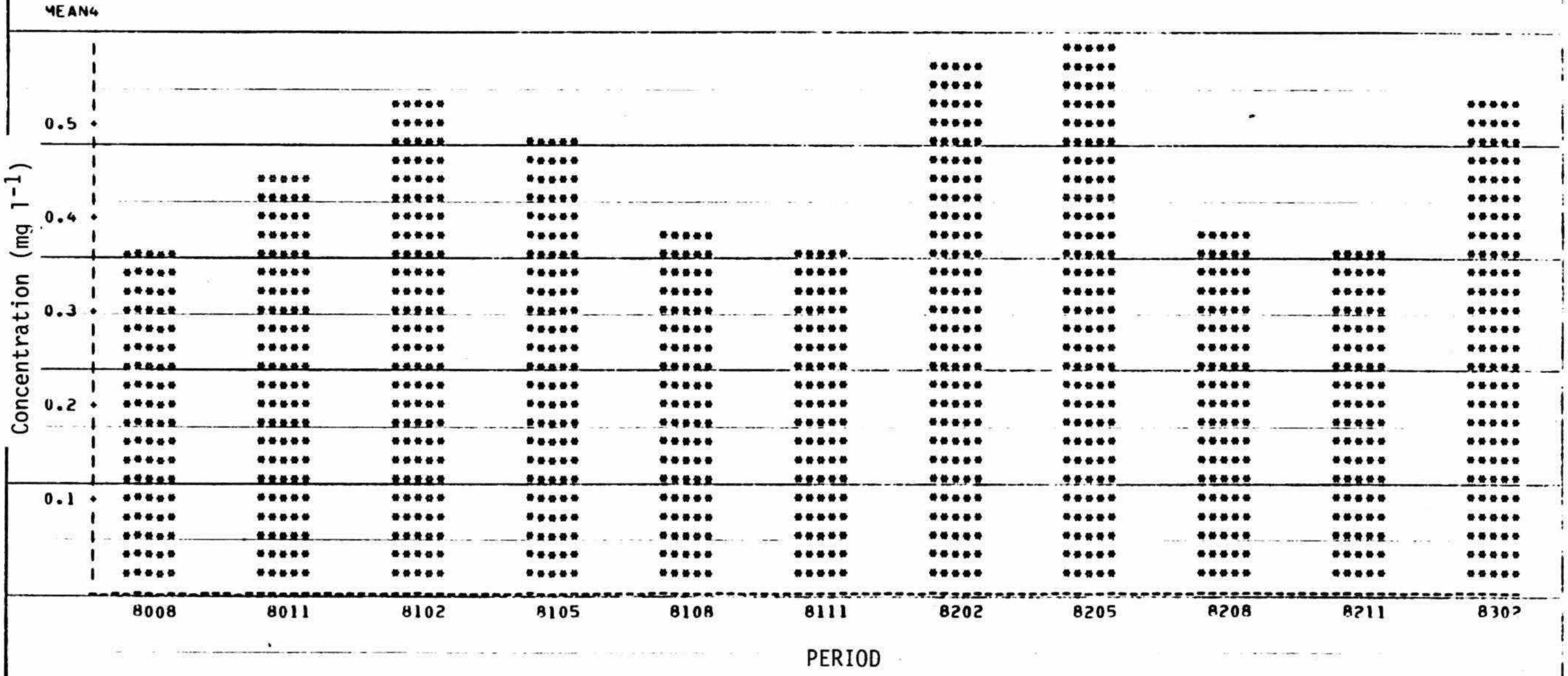
STATISTICAL ANALYSIS SYSTEM

FIGURE 4-3 : BAR CHART OF MEAN3 ($SO_4^=$)



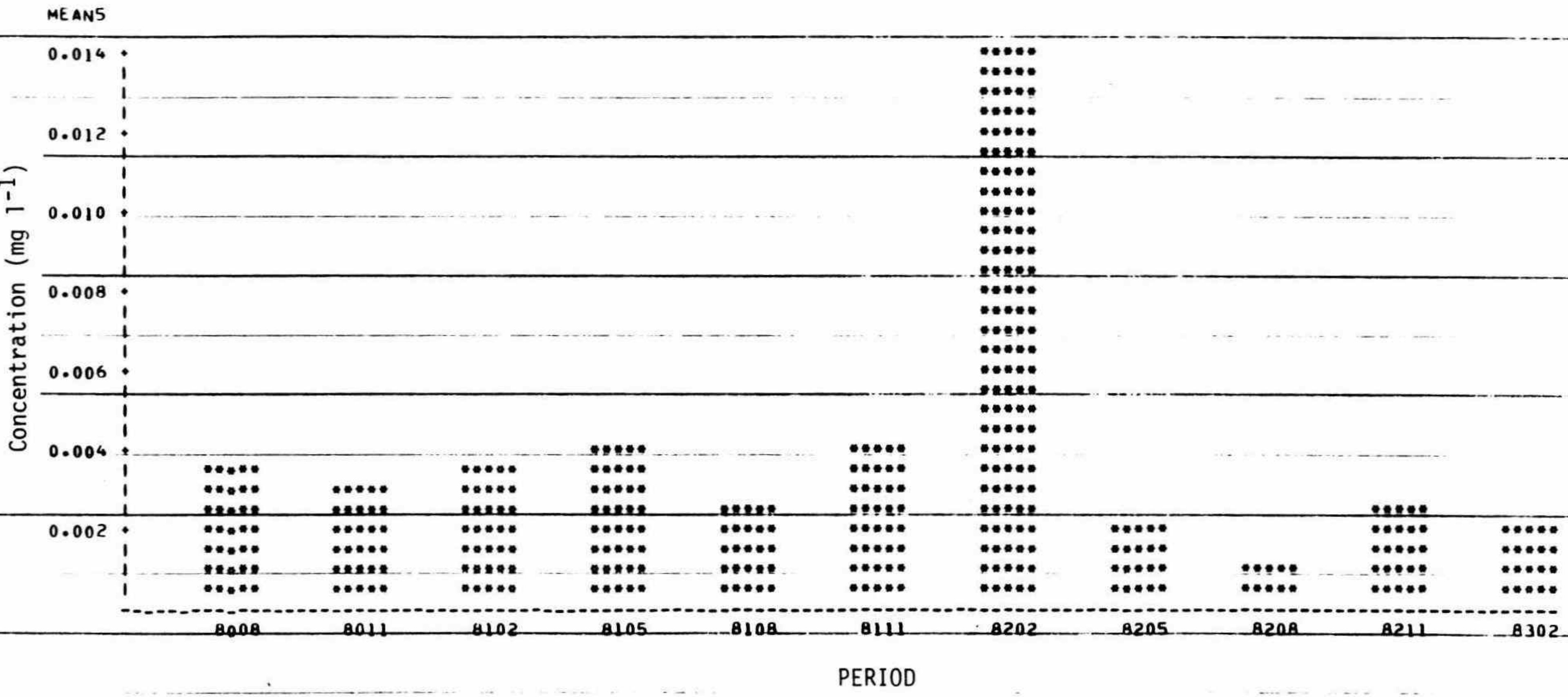
STATISTICAL ANALYSIS SYSTEM

FIGURE 4-4 : BAR CHART OF MEAN4 ($N\text{-}NO_3^-$)



STATISTICAL ANALYSIS SYSTEM

FIGURE 4-5 : BAR CHART OF MEANS (Cu)



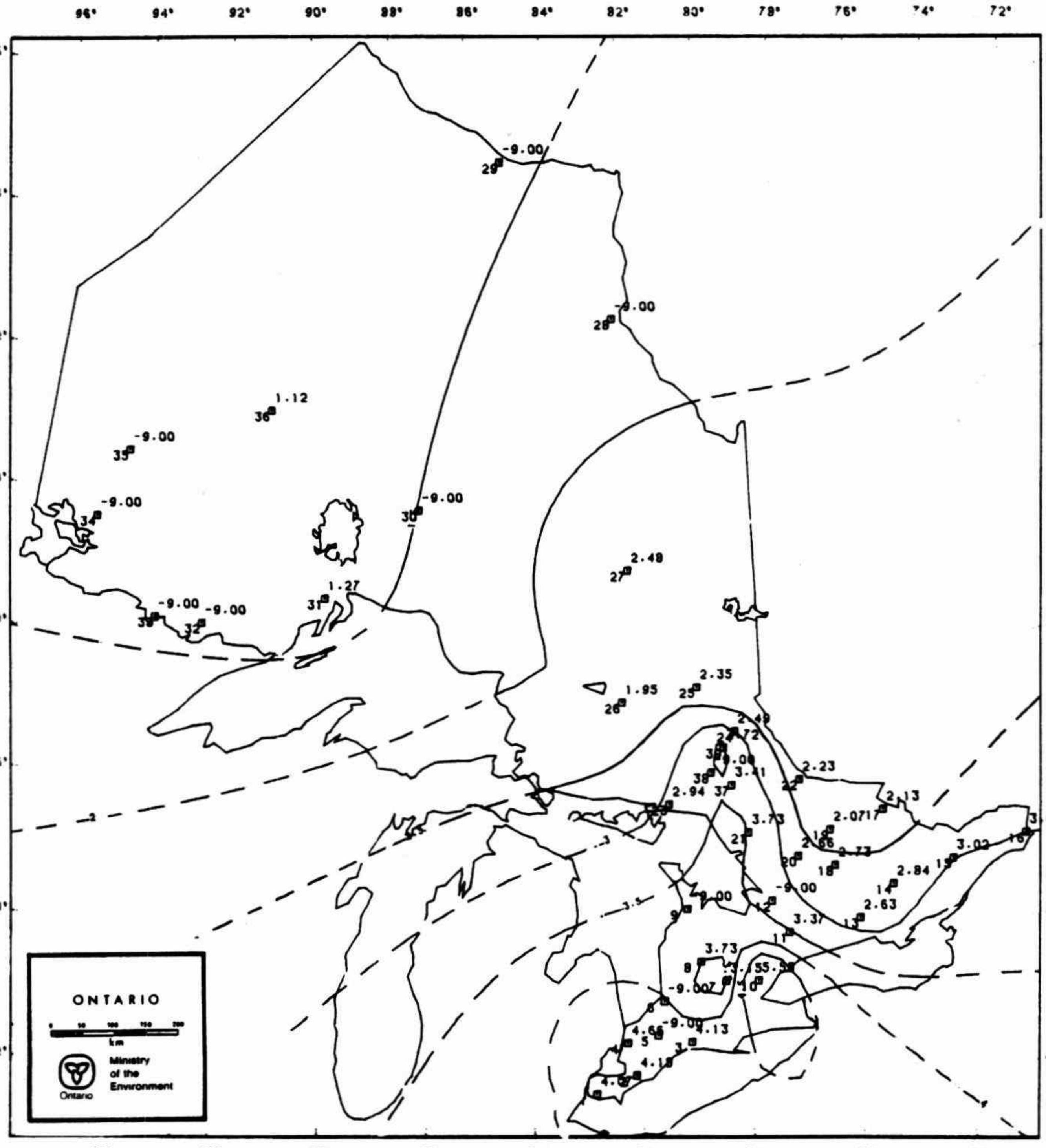


FIGURE 5-1:SO4 CONCENTRATION (MG/L) FROM JULY 1980 TO MARCH 1981

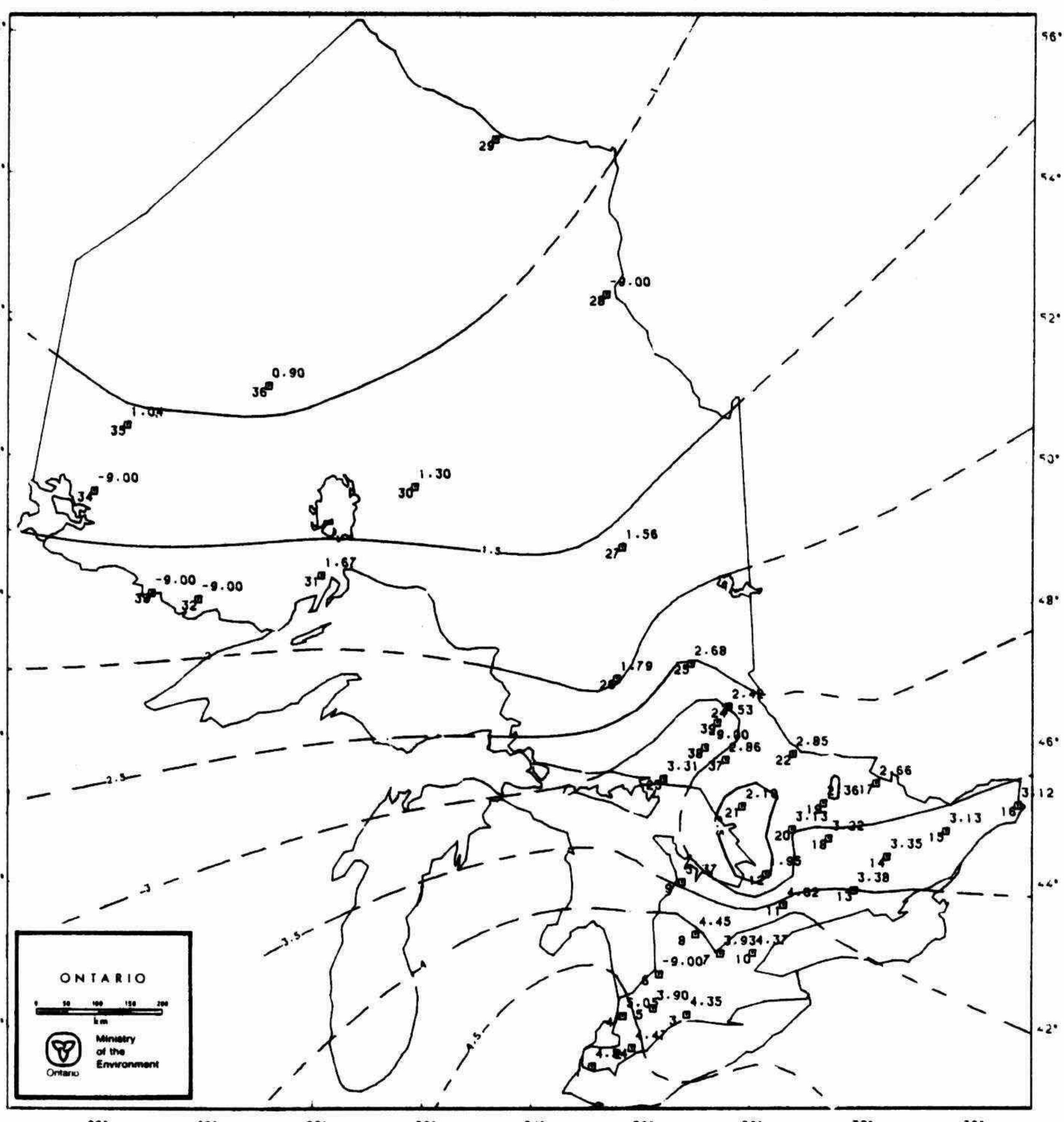


FIGURE 5-2:SO4 CONCENTRATION (MG/L) FROM JULY 1981 TO MARCH 1982

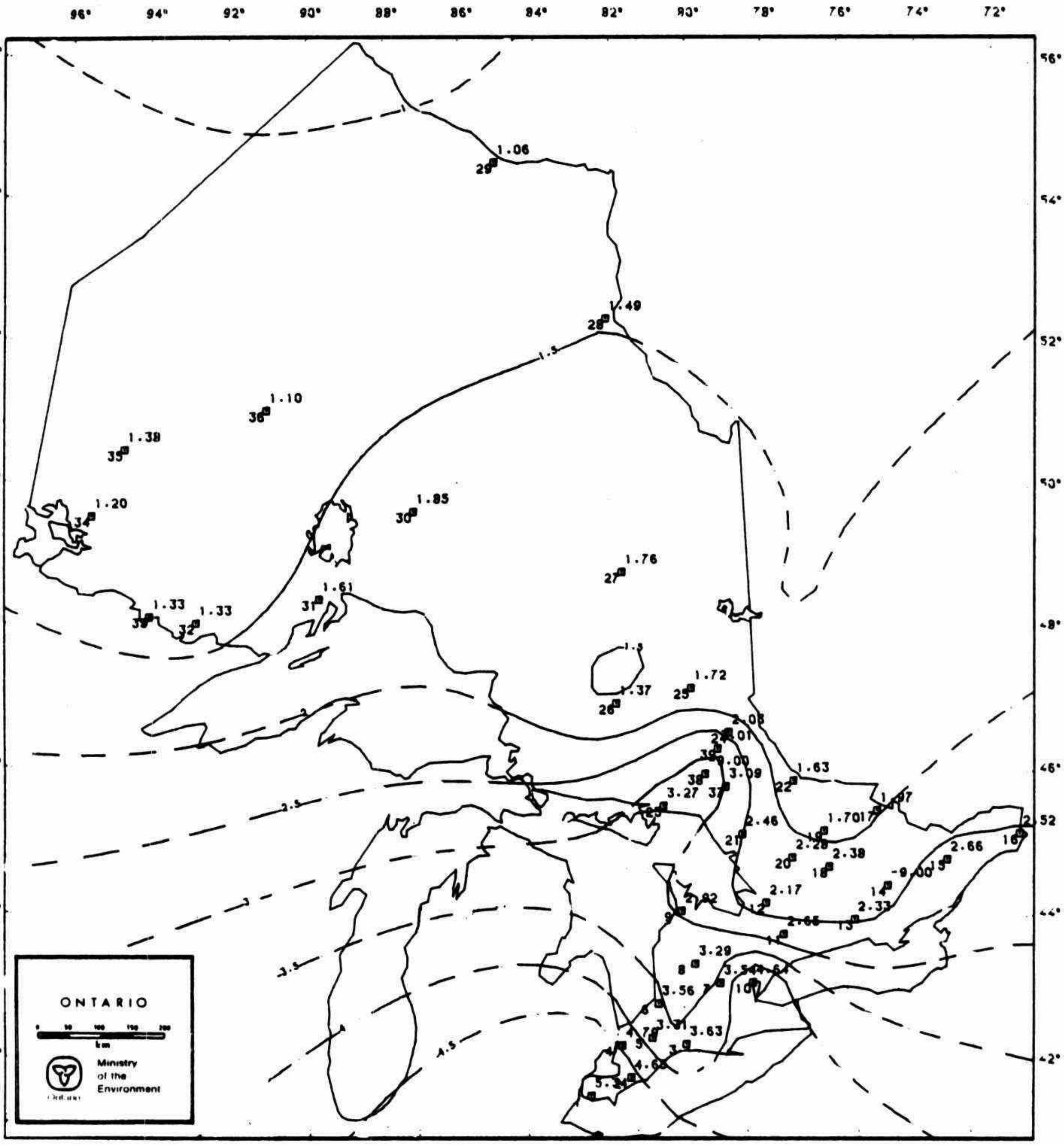


FIGURE 5-3: SO4 CONCENTRATION (MG/L) FROM JULY 1982 TO MARCH 1983

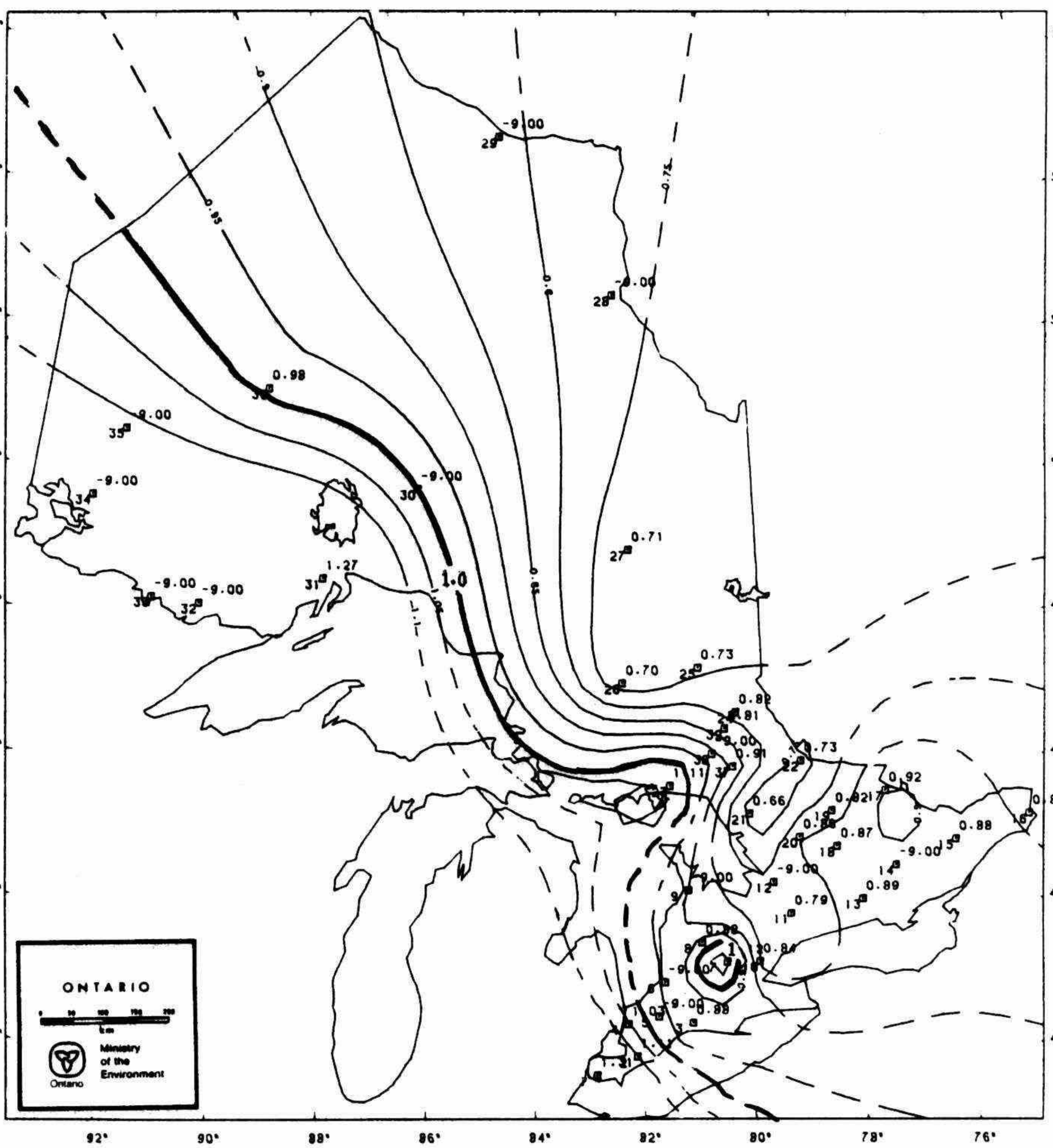


FIGURE 5-4: RATIO OF SO4 CONCENTRATION BETWEEN 82-83 AND 80-81

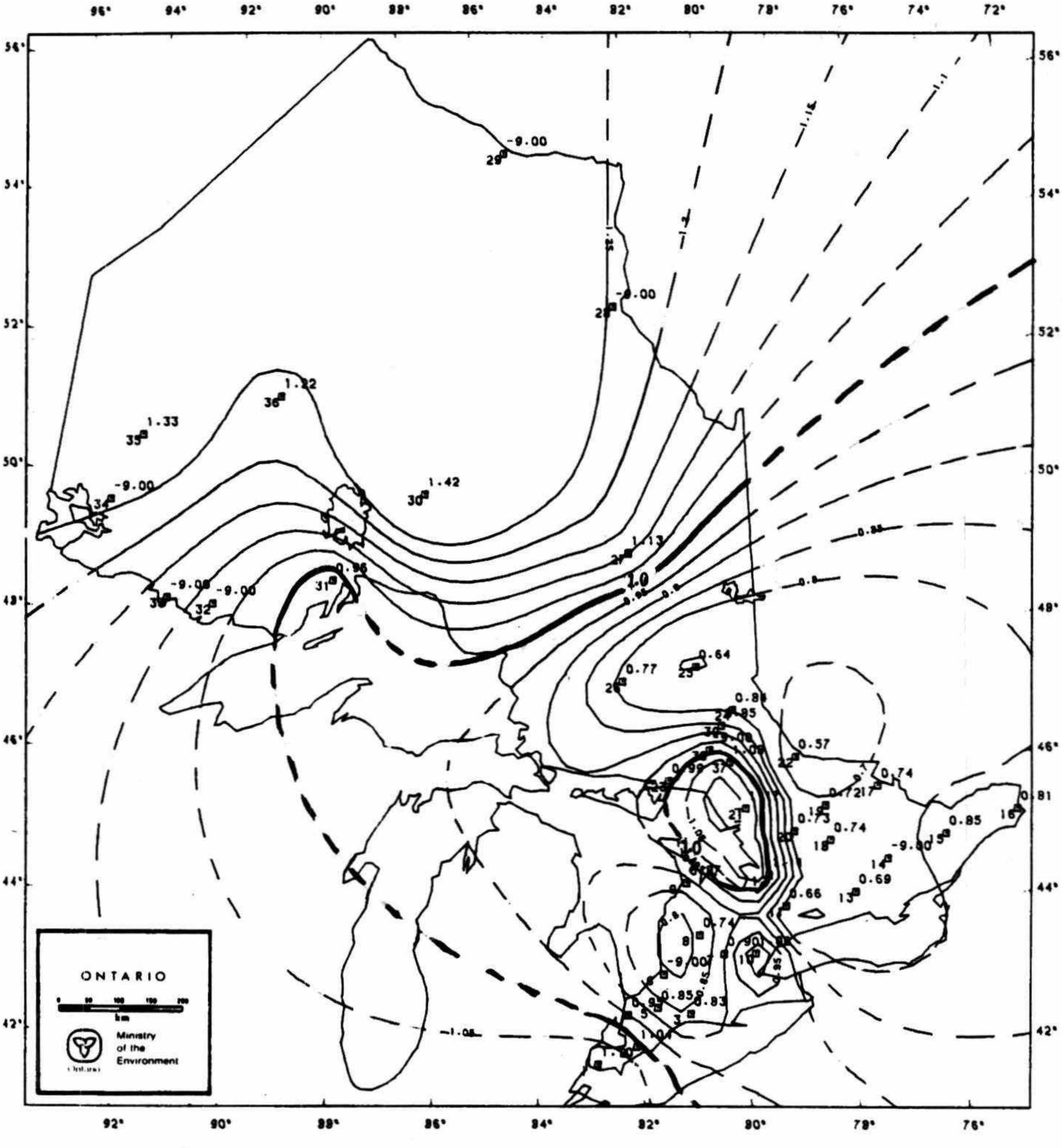


FIGURE 5-5 RATIO OF SO4 CONCENTRATION BETWEEN 82-83 AND 81-82

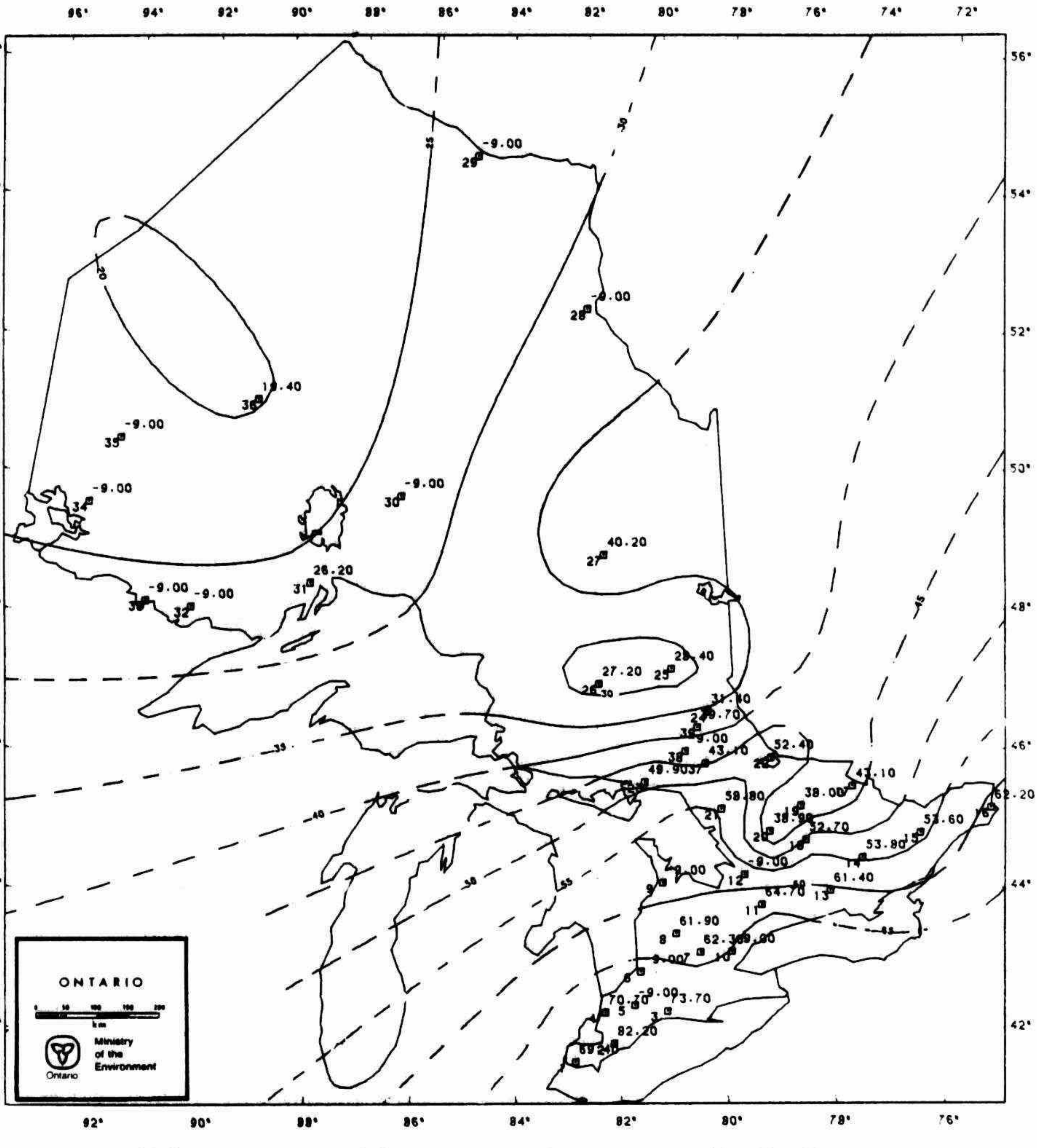


FIGURE 5-6:N-NO3 CONCENTRATION (MG*100/L) FROM JULY 1980 TO MAR 1981

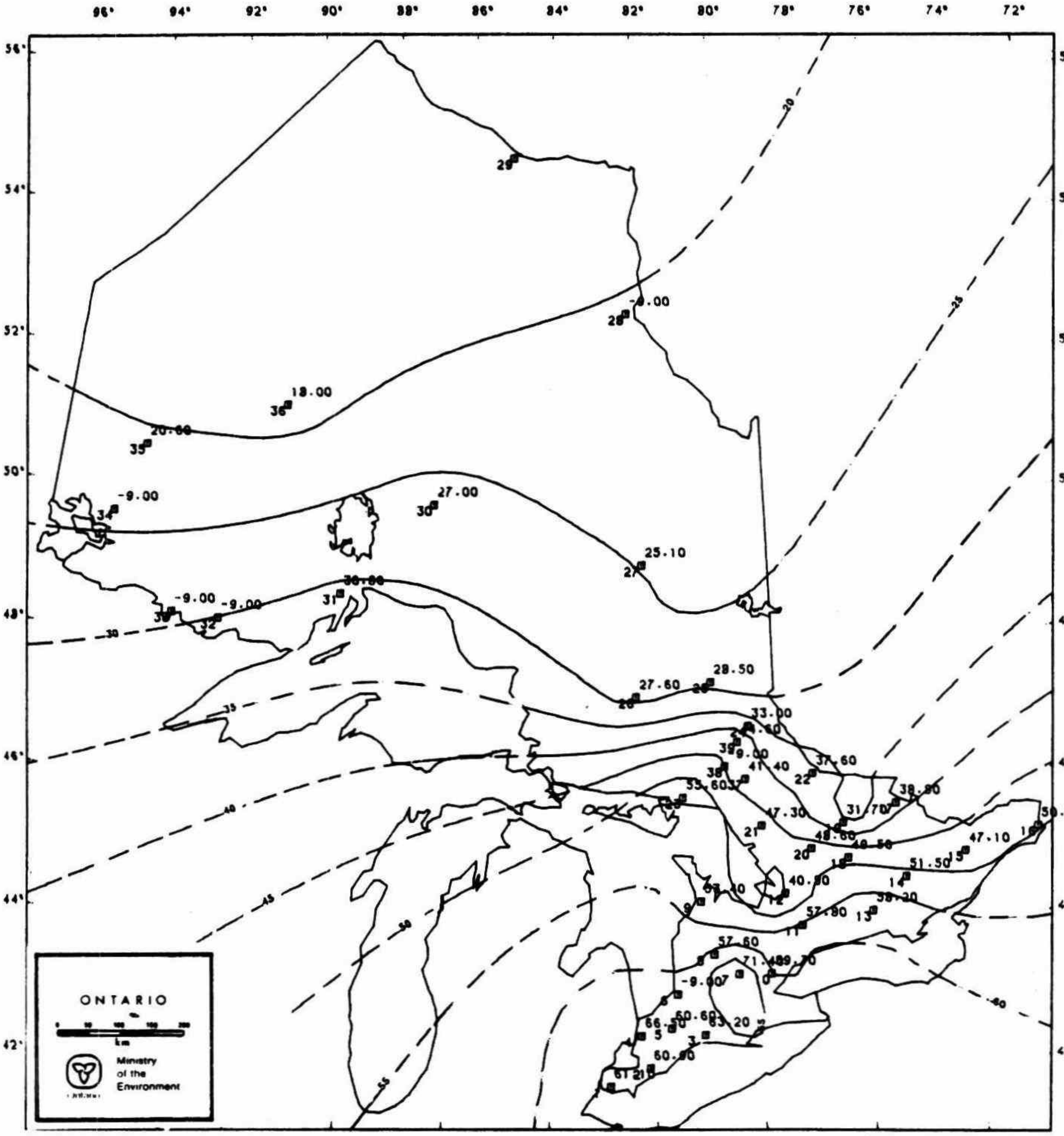


FIGURE 5-7: N-NO3 CONCENTRATION (MG•100/L) FROM JULY 1981 TO MAR 1982

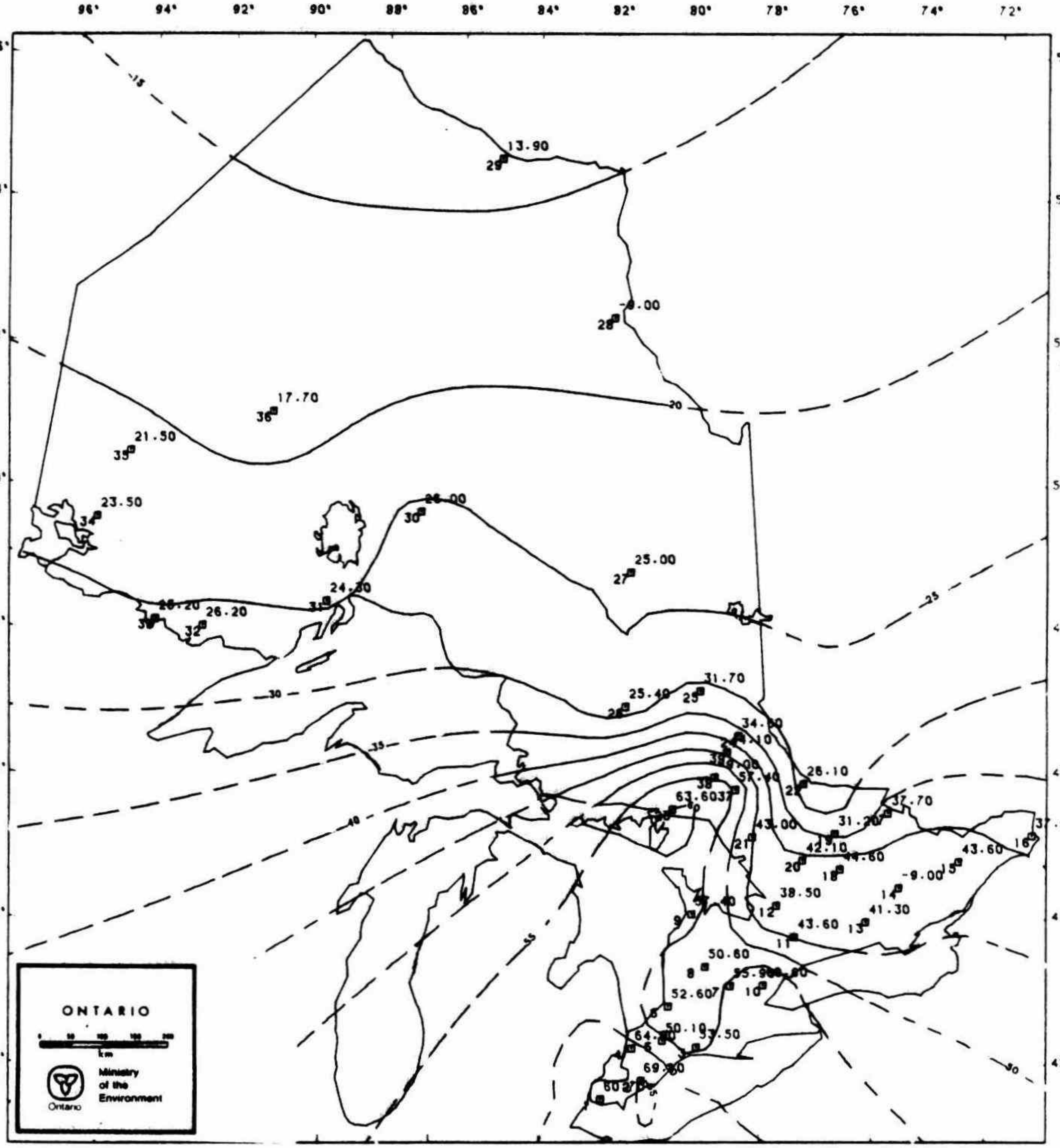


FIGURE 5-8: N-NO3 CONCENTRATION (MG•100/L) FROM JULY 1982 TO MAR 1983

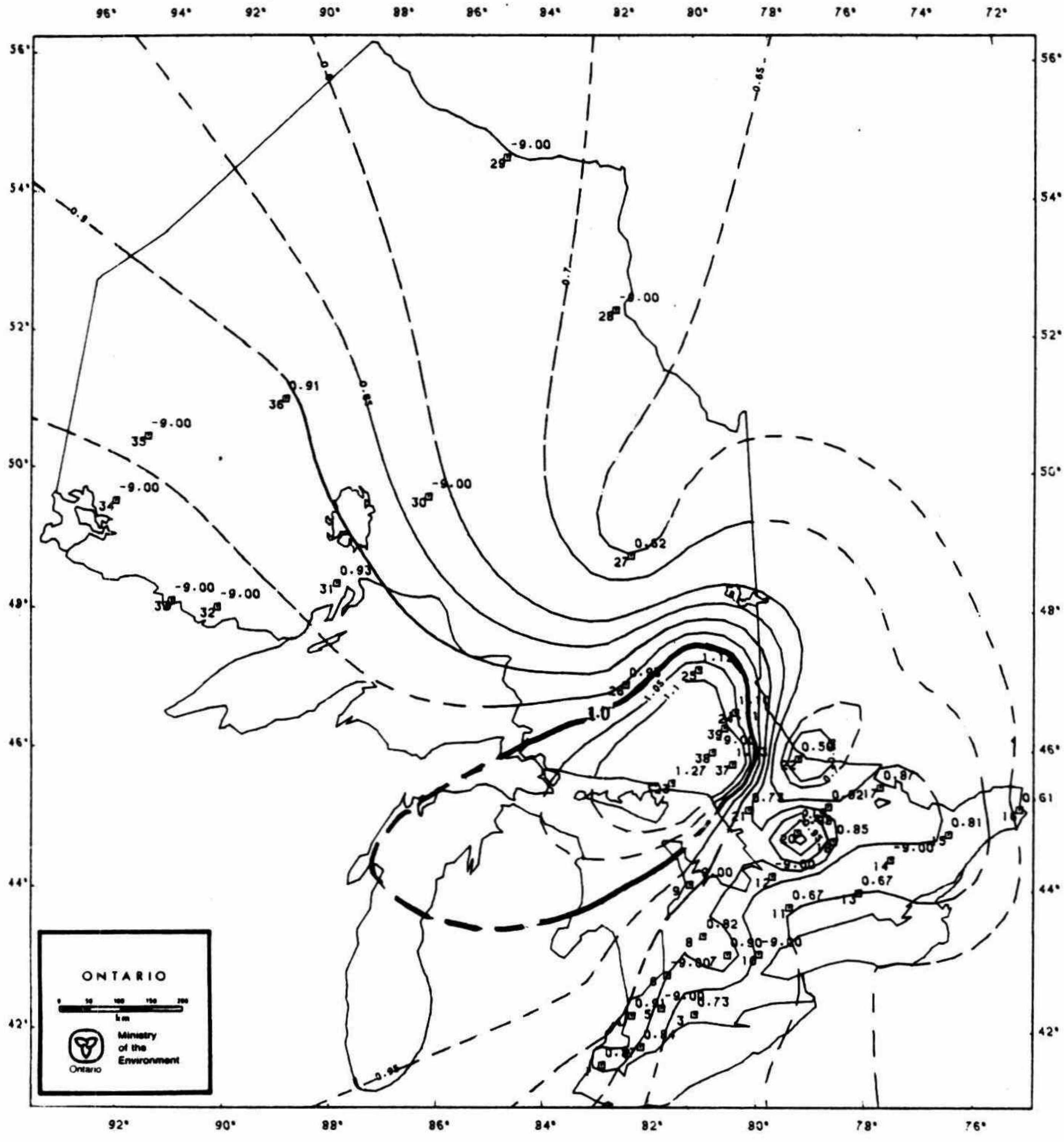


FIGURE5-9, RATIO OF N-NO3 CONCENTRATION BETWEEN 82-83 AND 80-81

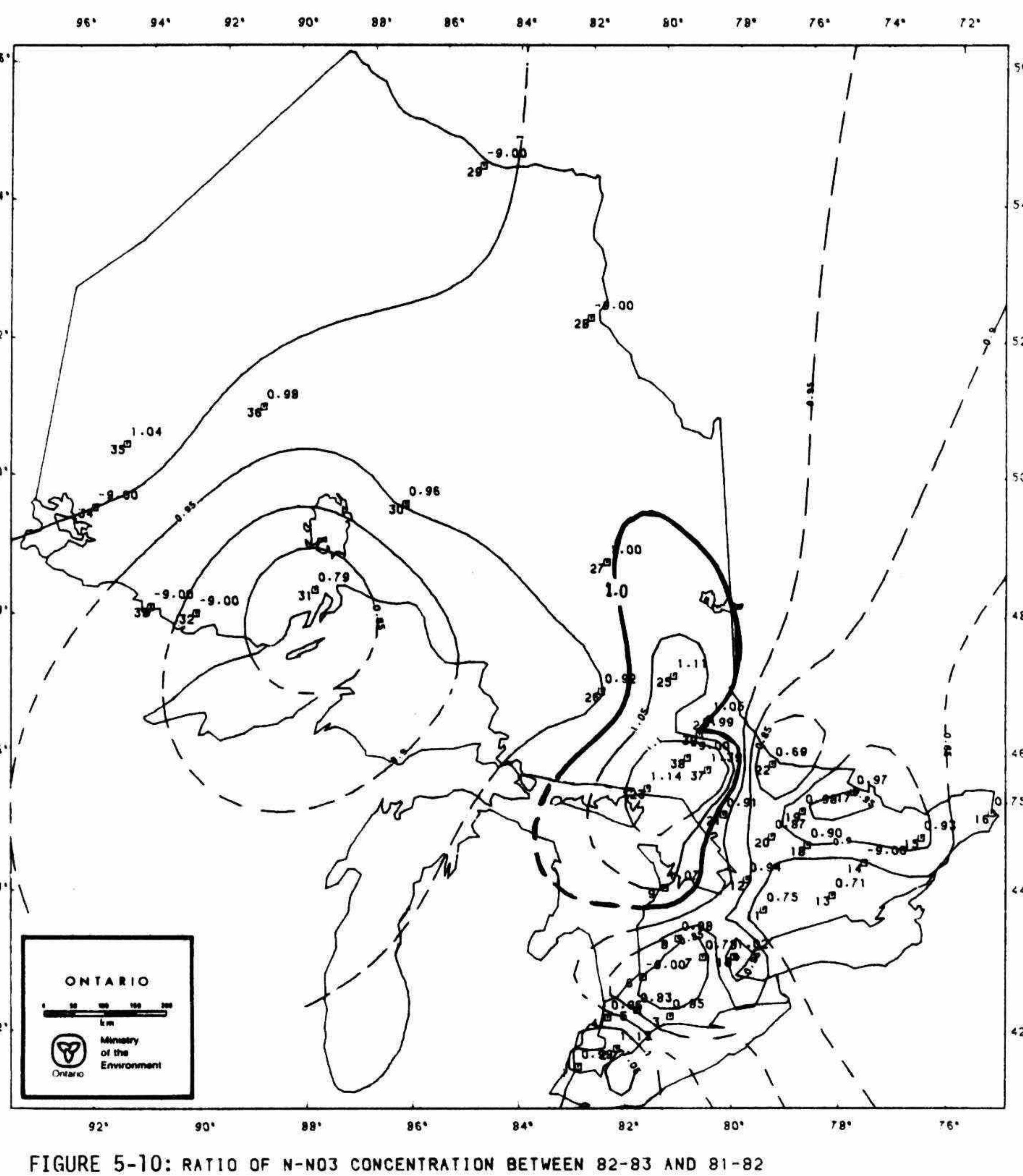


FIGURE 5-10: RATIO OF N-NO3 CONCENTRATION BETWEEN 82-83 AND 81-82

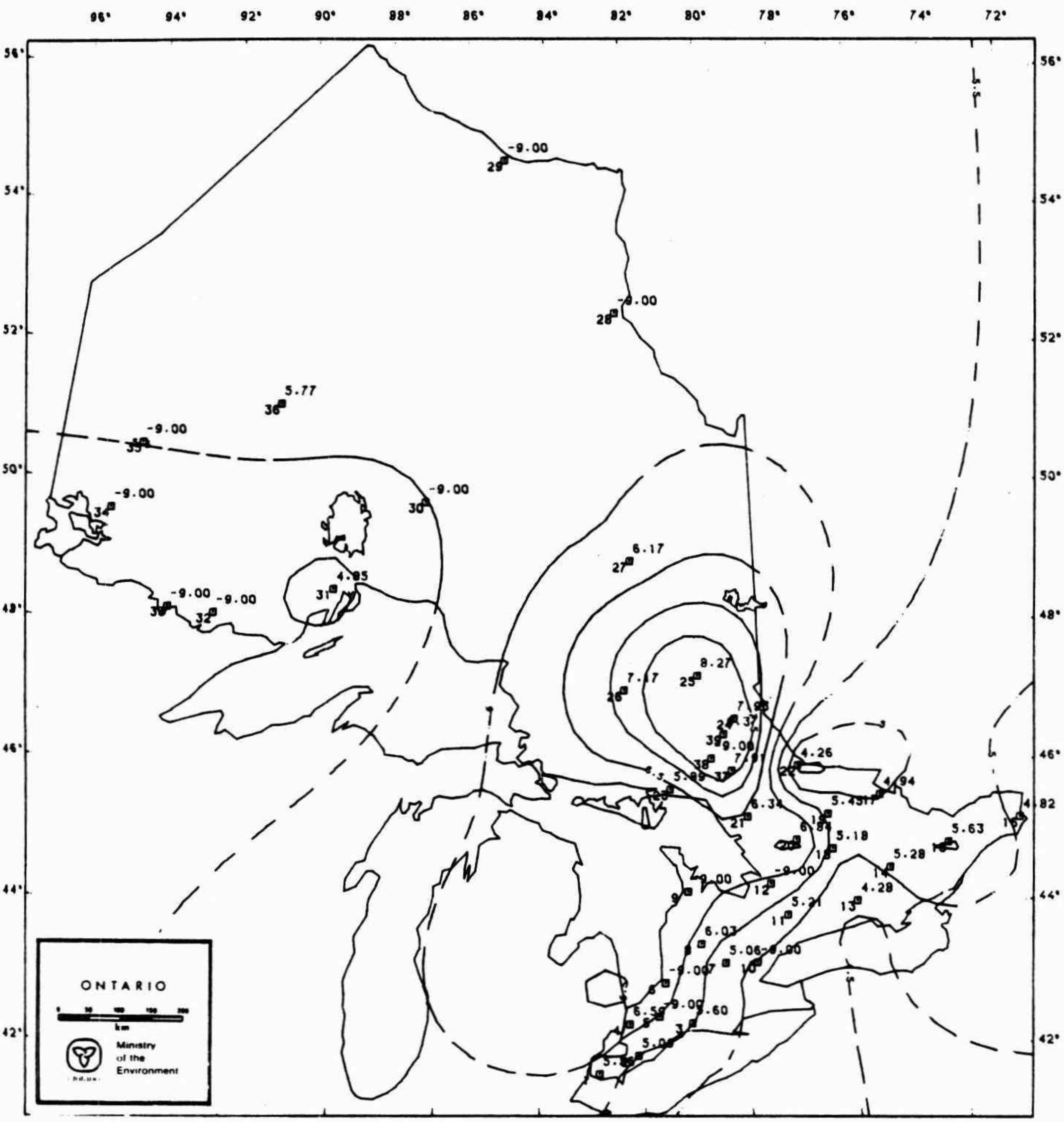


FIGURE 5-11: SO4/N-NO3 RATIO FROM JULY 1980 TO MAR 1981

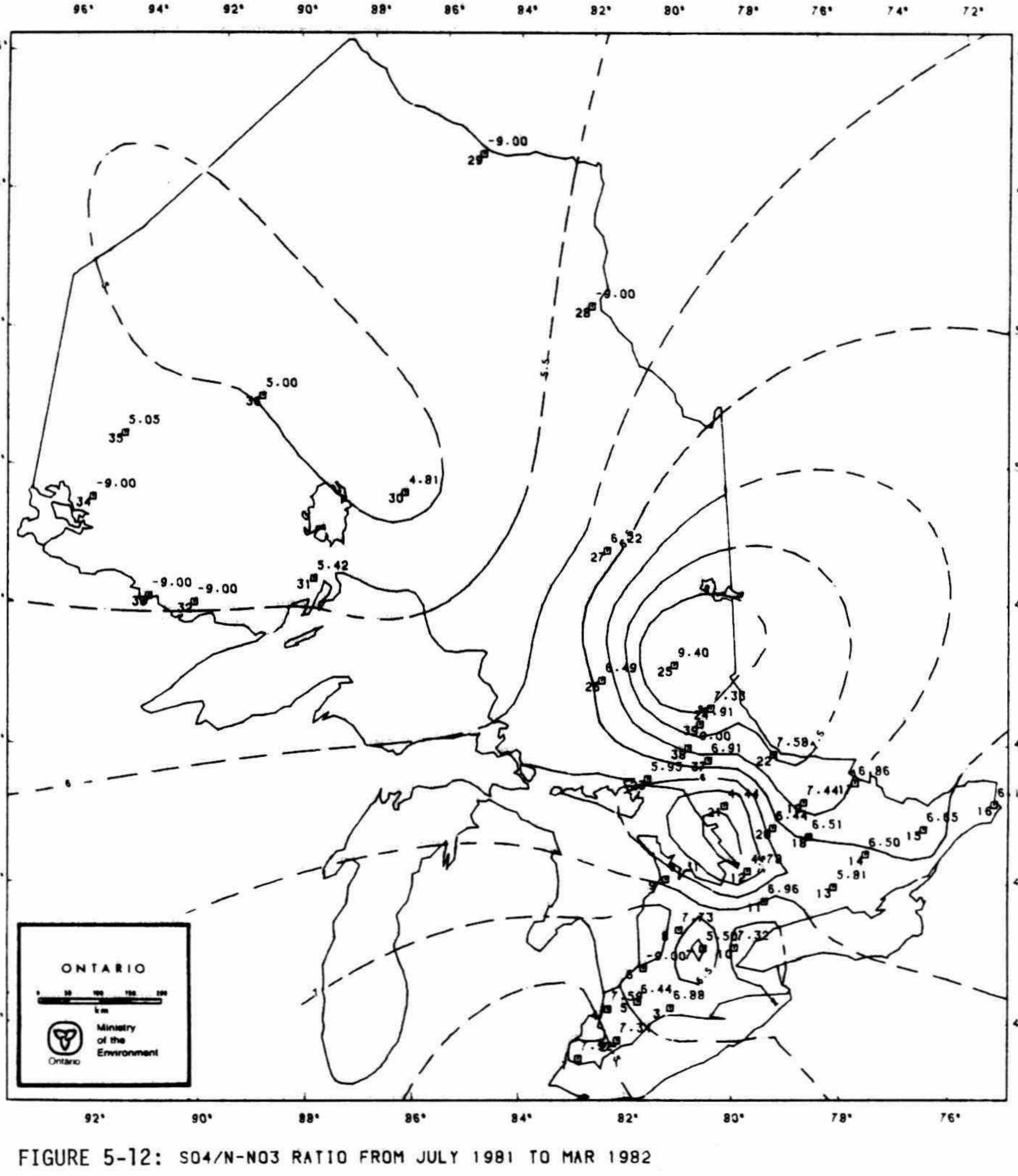


FIGURE 5-12: SO4/N-NO3 RATIO FROM JULY 1981 TO MAR 1982

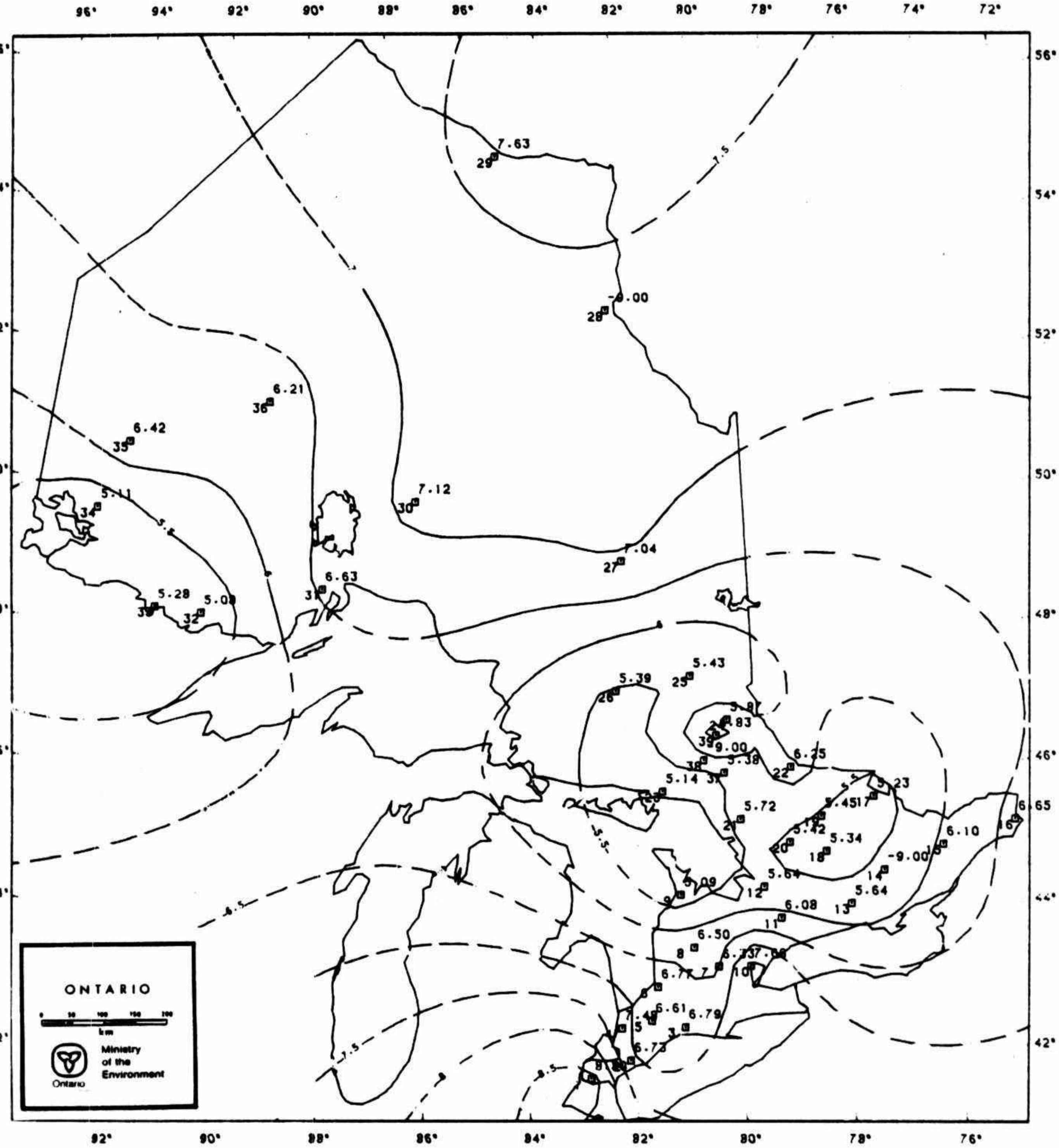


FIGURE 5-13: SO4/N-NO3 RATIO FROM JULY 1982 TO MAR 1983

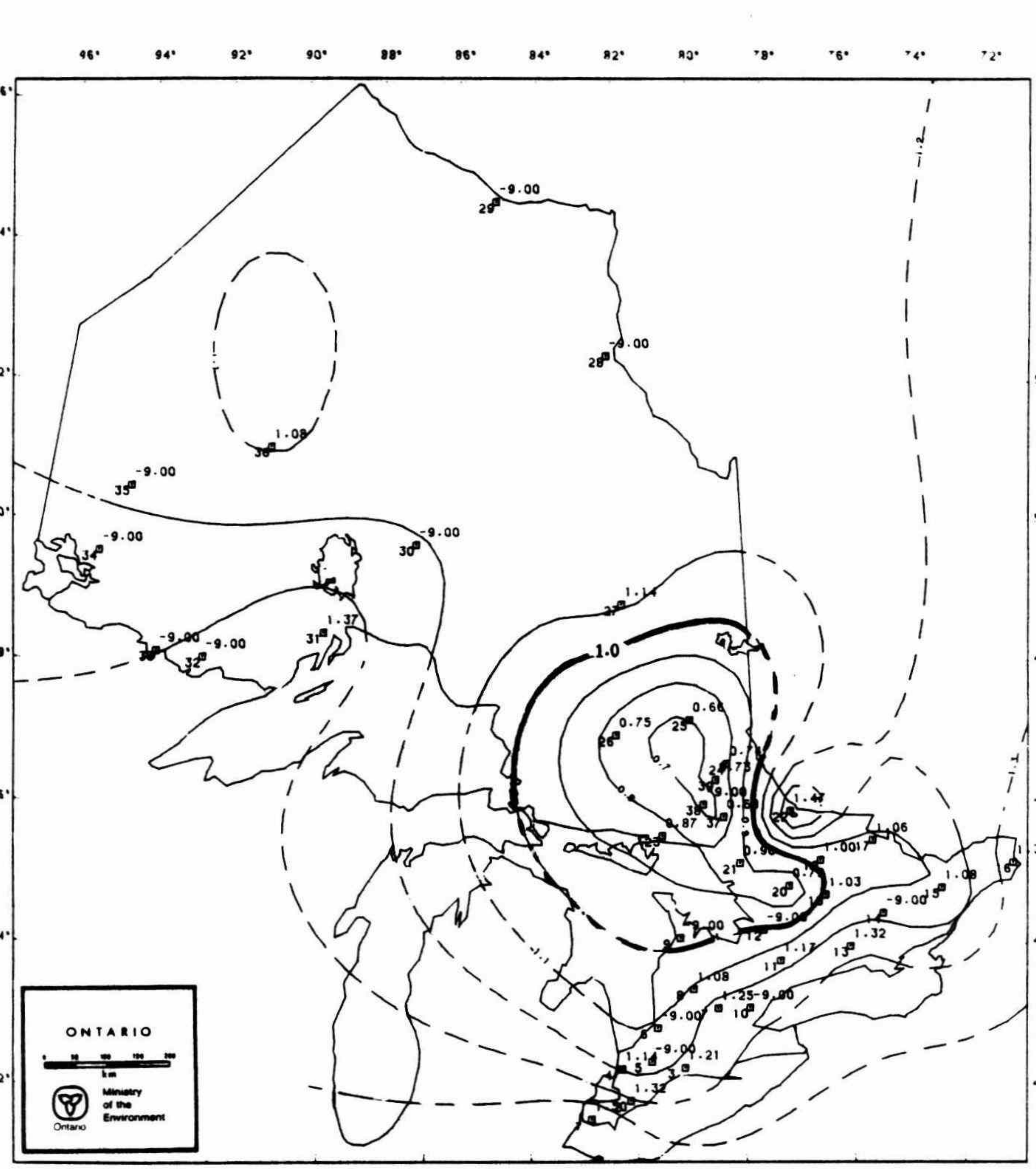


FIGURE 5-14: RATIO OF SO4/N-NO3 RATIOS OF 82-83 AND 80-81

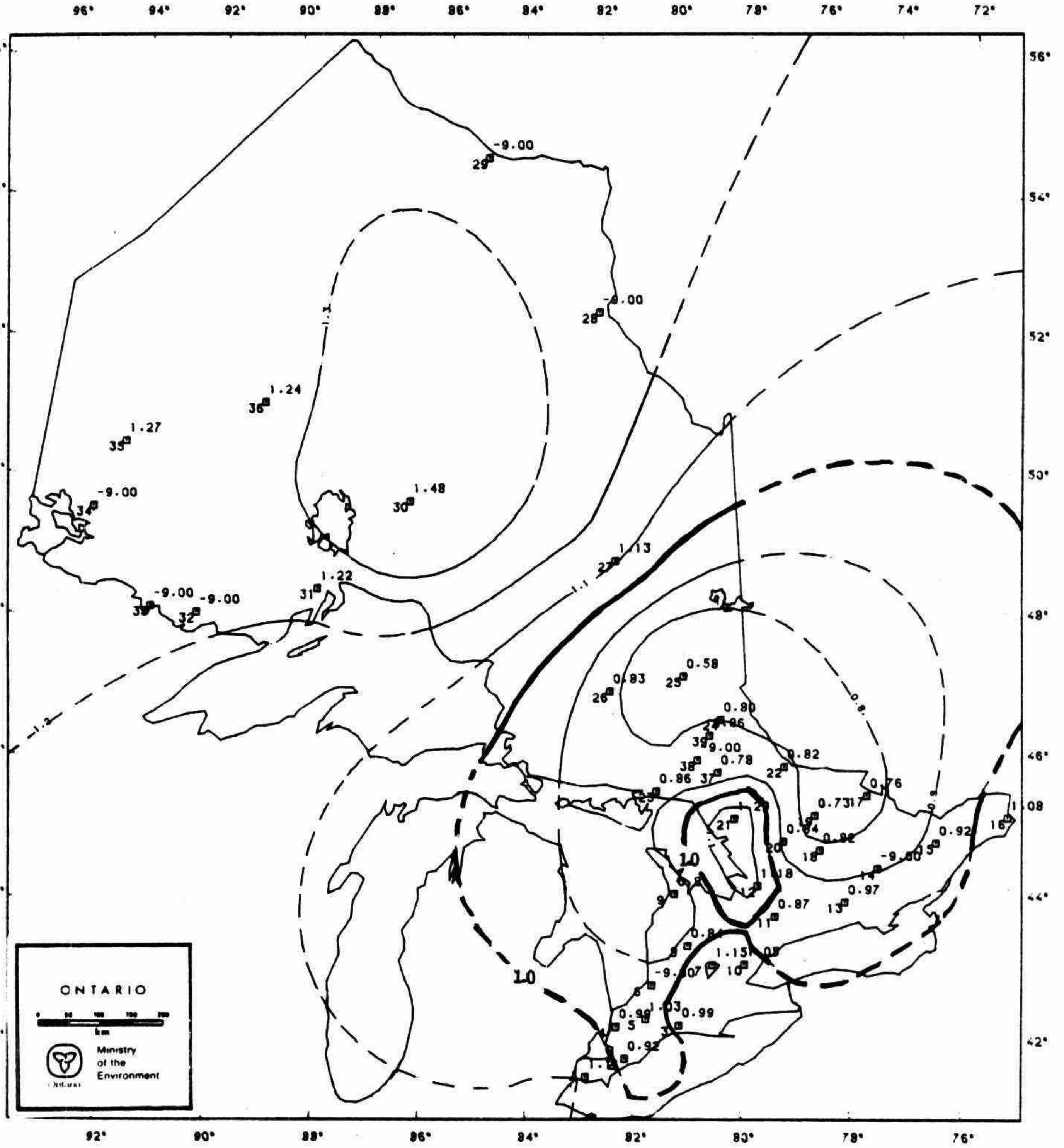


FIGURE 5-15: RATIO OF SO4/N-NO3 RATIOS OF 82-83 AND 81-82

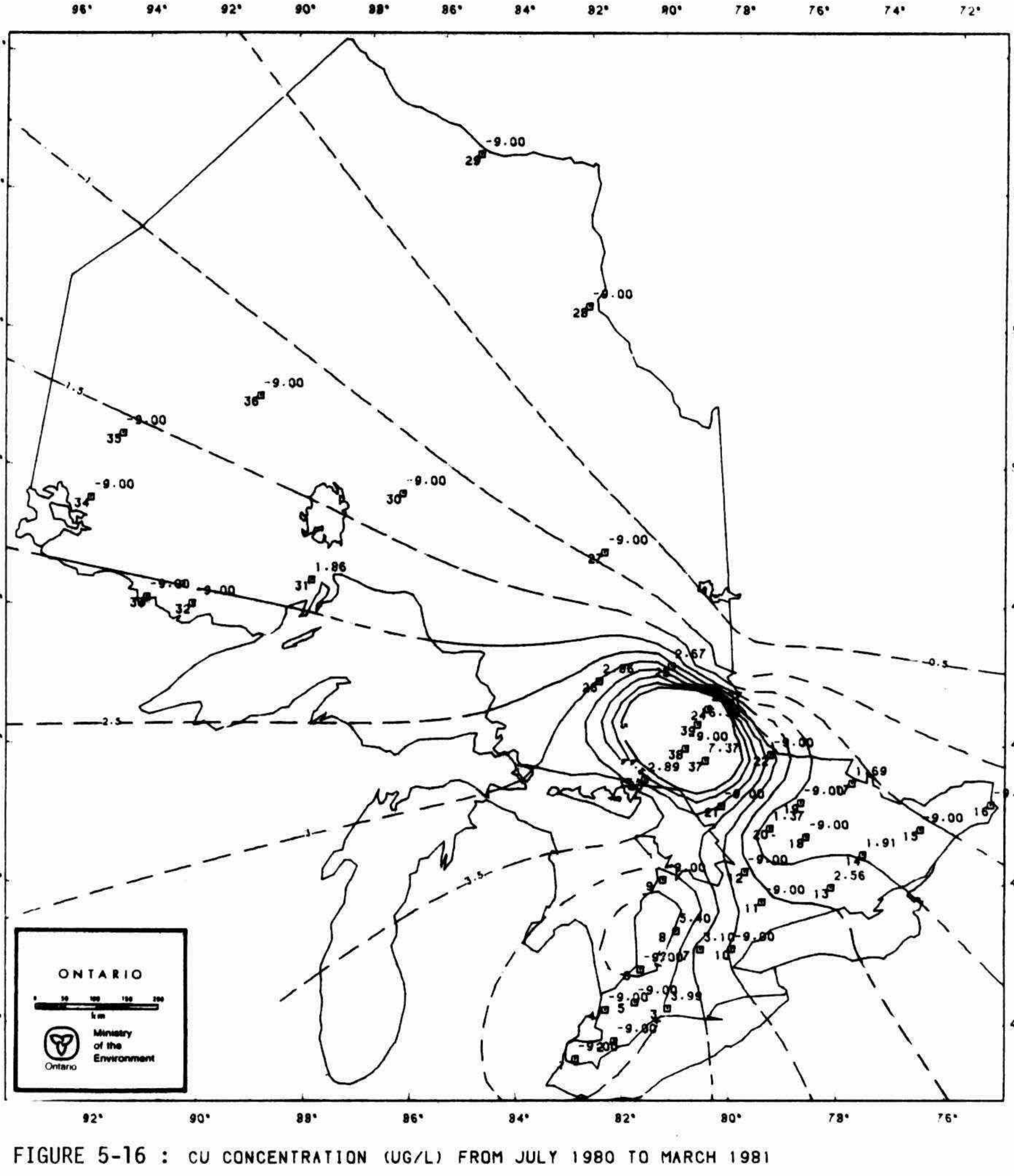


FIGURE 5-16 : CU CONCENTRATION (UG/L) FROM JULY 1980 TO MARCH 1981

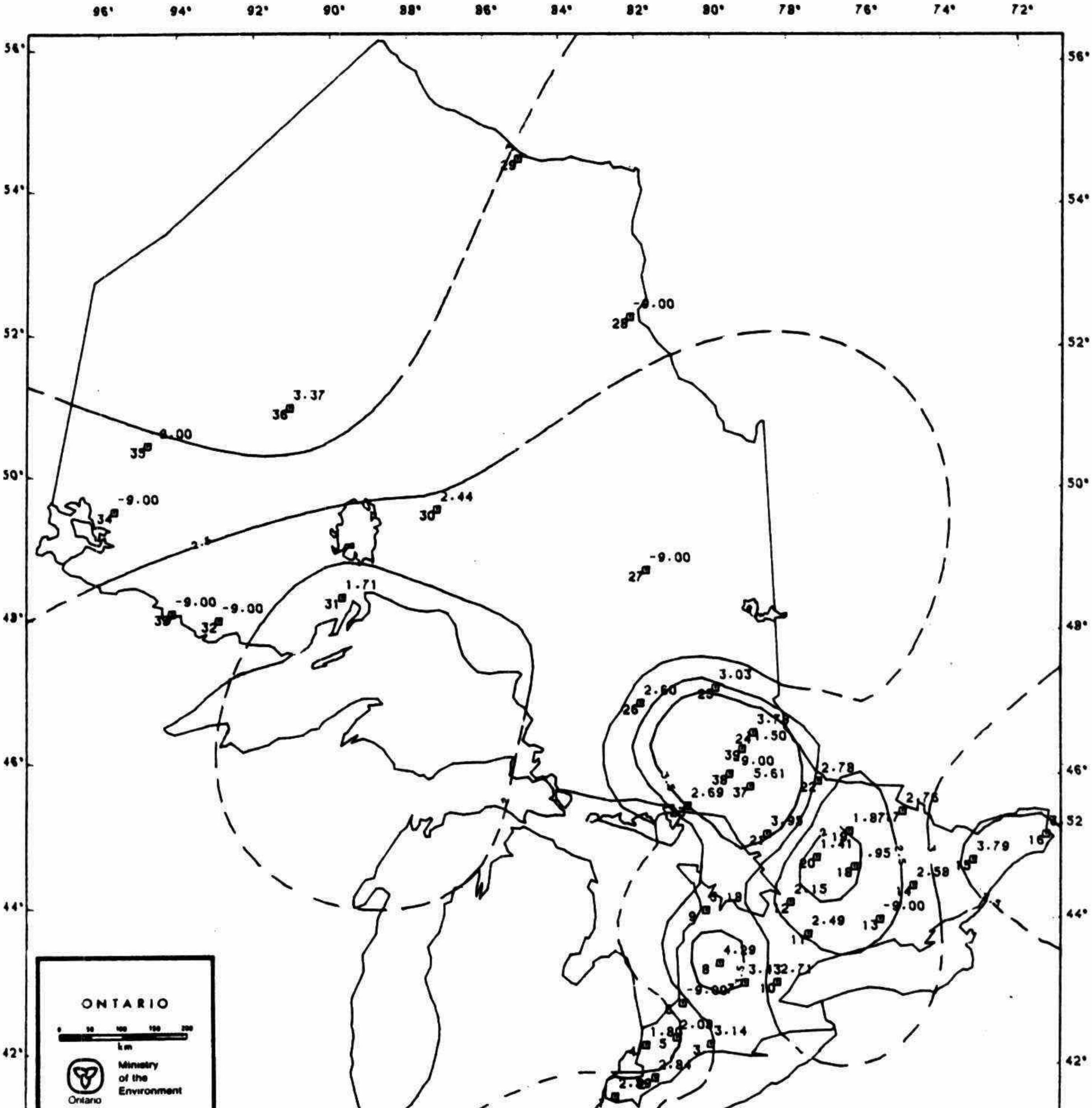


FIGURE 5-17 : CU CONCENTRATION (UG/L) FROM JULY 1981 TO MARCH 1982

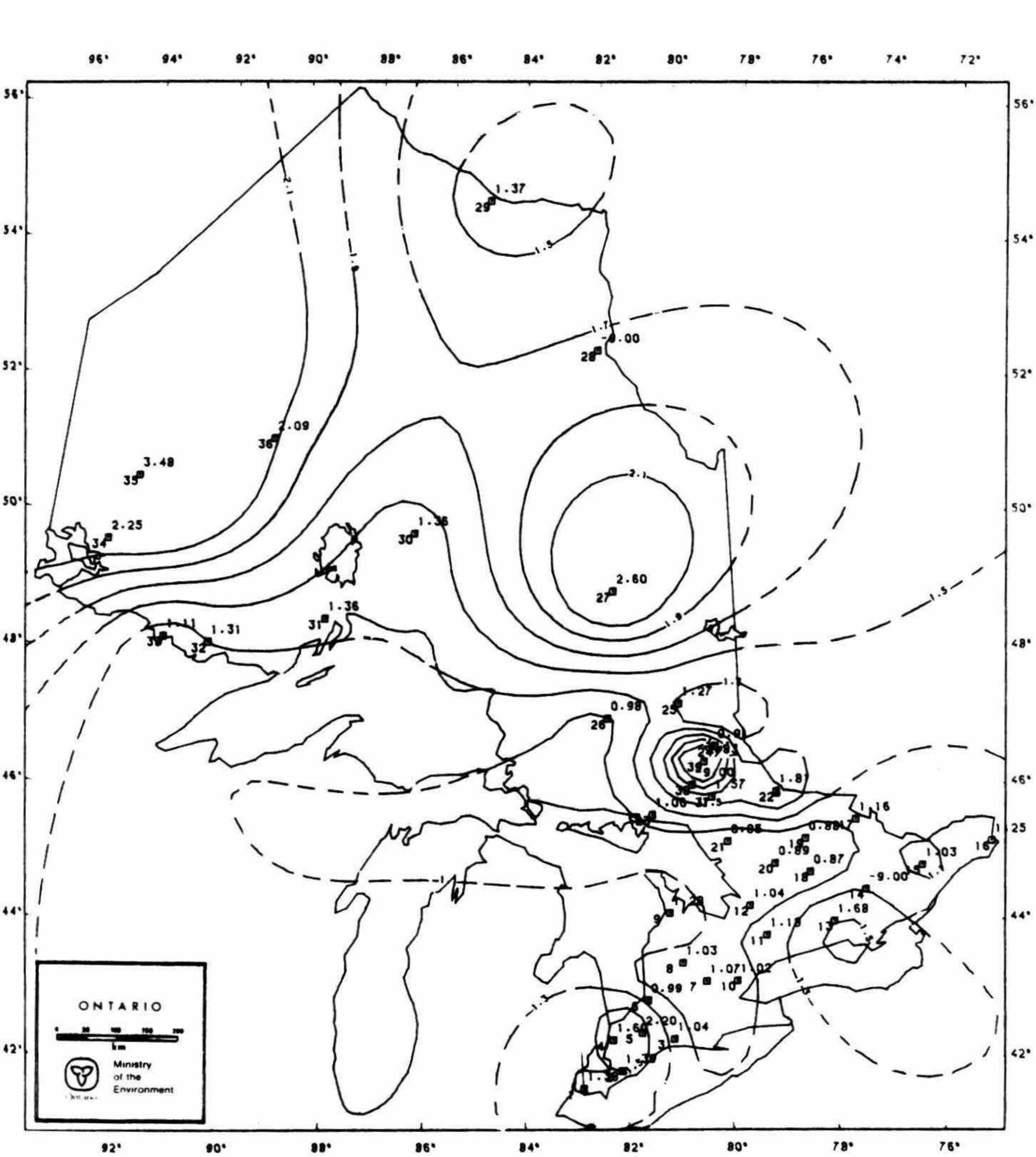


FIGURE 5-18 : CU CONCENTRATION (UG/L) FROM JULY 1982 TO MARCH 1983

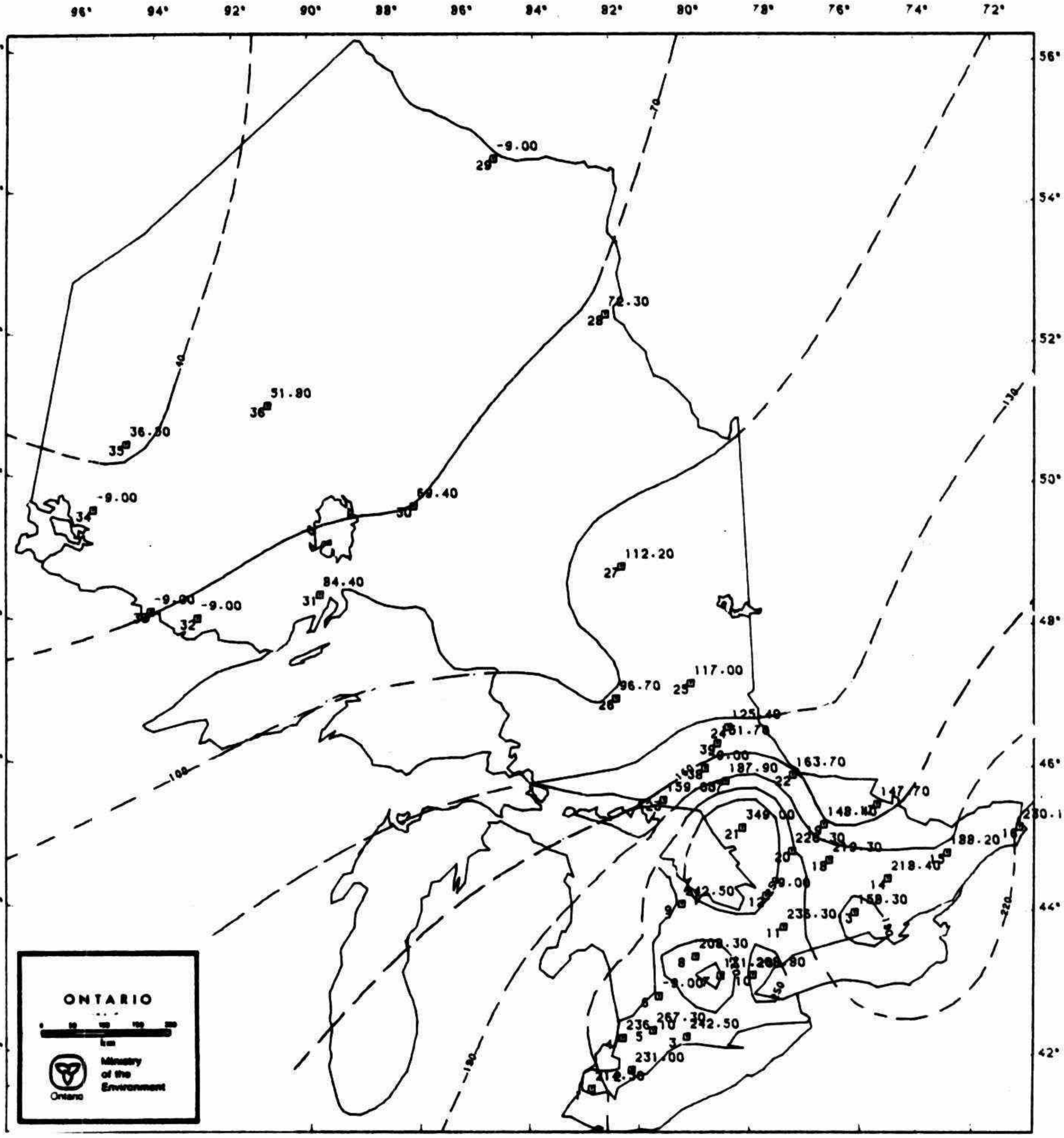


FIGURE 5-19 : SO4 DEPOSITION (MG•10/M••2) FROM JULY 1980 TO MAR 1981

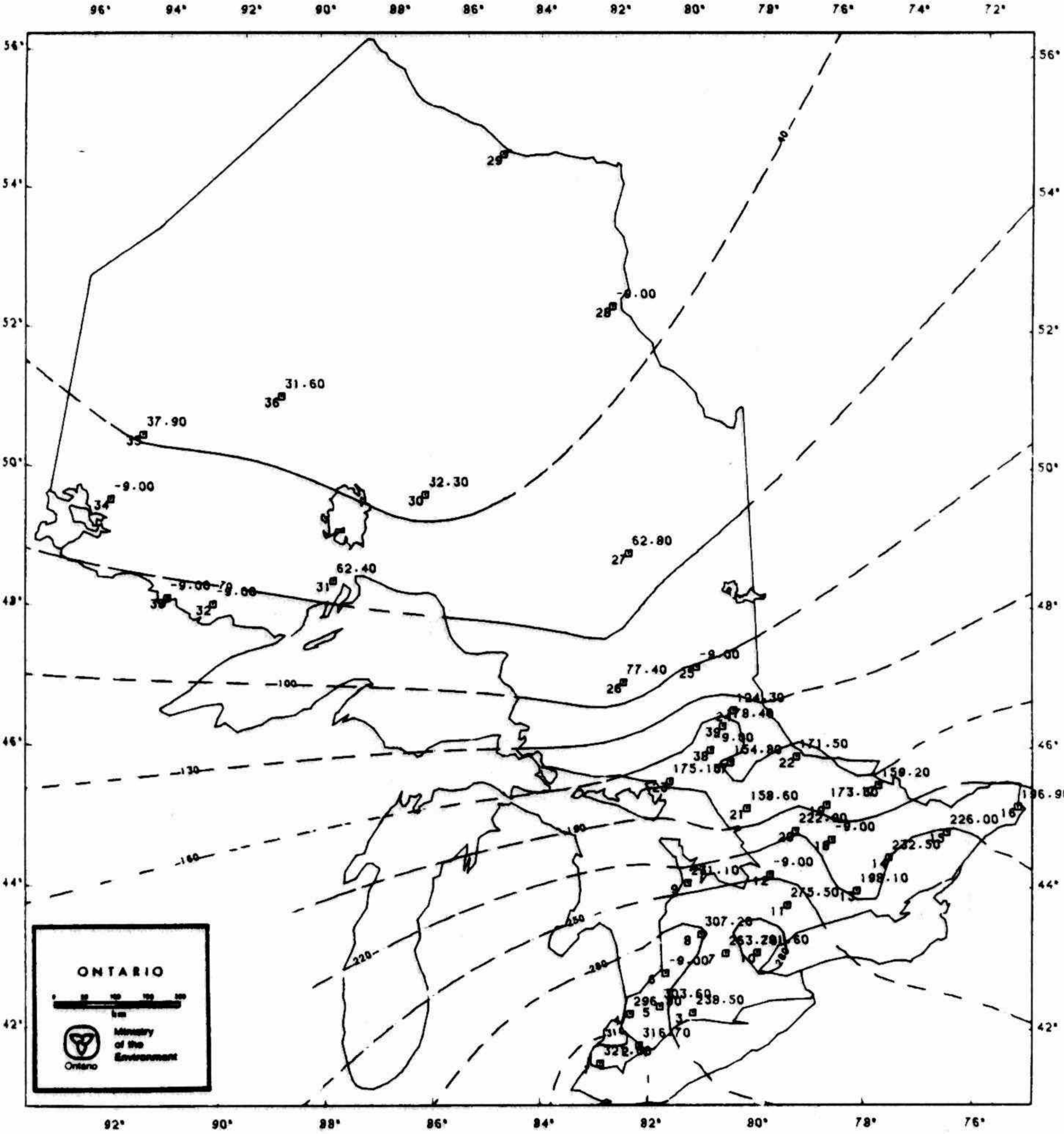


FIGURE 5-20 : SO4 DEPOSITION (MG*10/M**2) FROM JULY 1981 TO MAR 1982

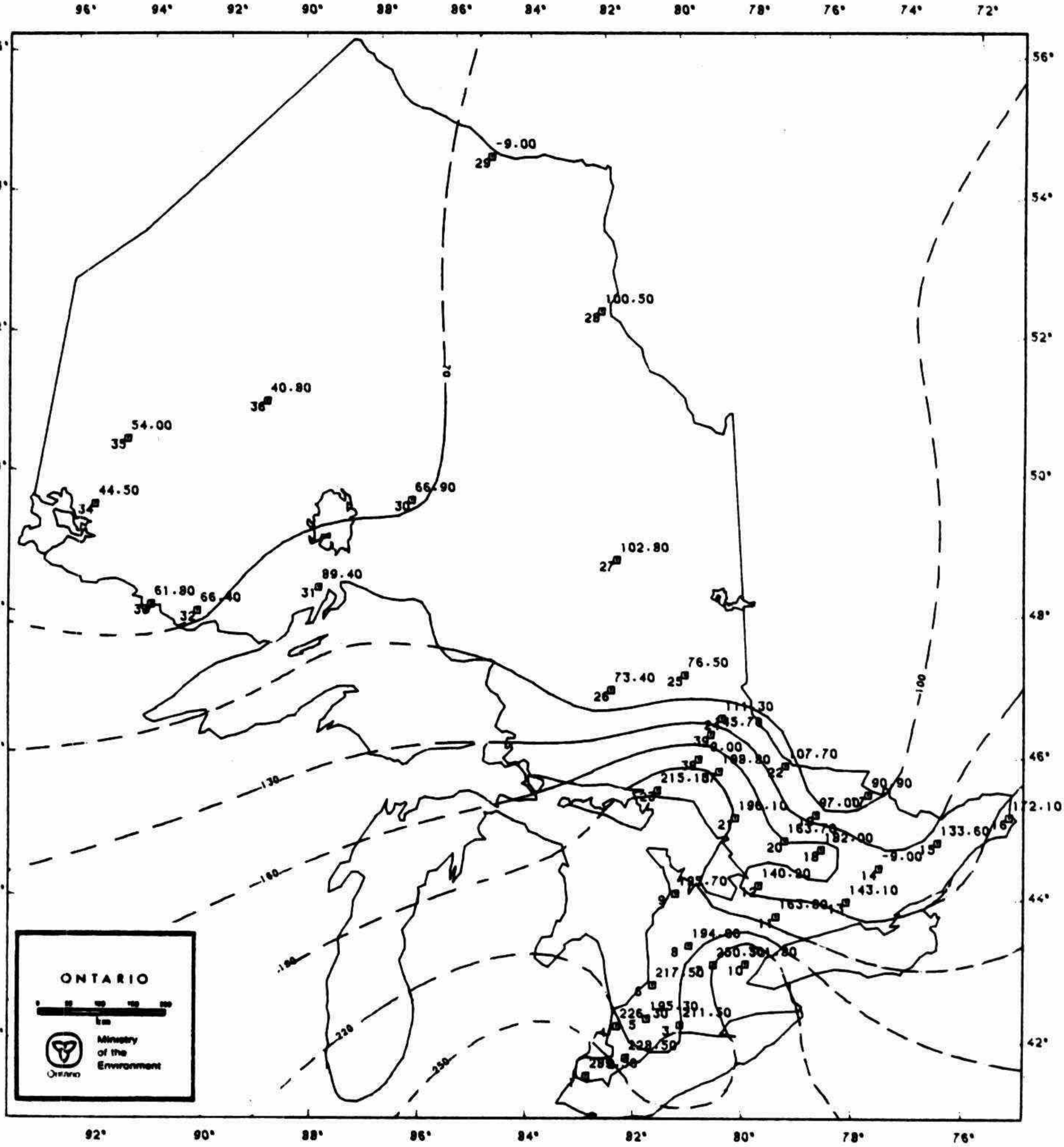


FIGURE 5-21 : SO4 DEPOSITION (MG*10/M**2) FROM JULY 1982 TO MAR 1983

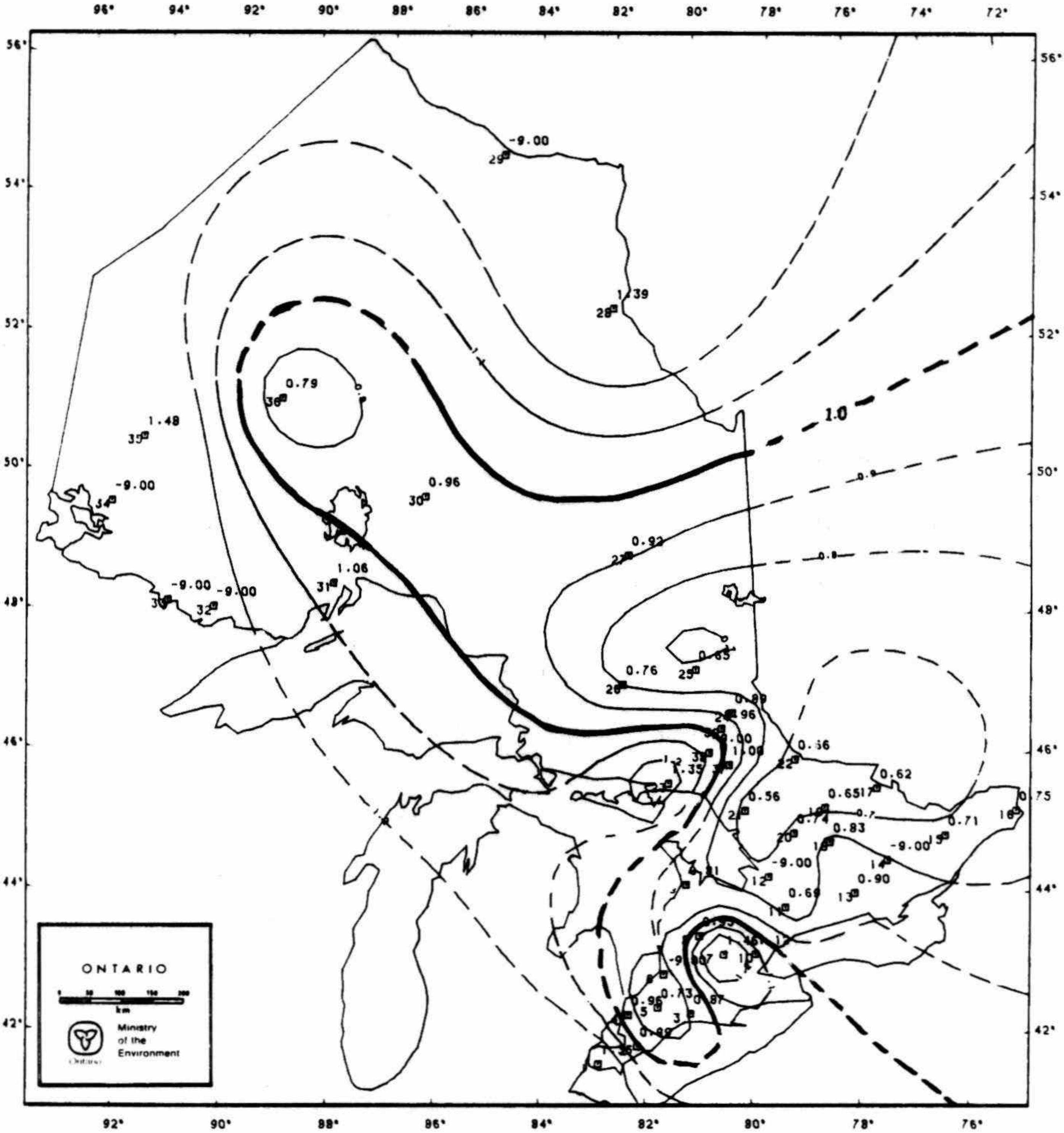


FIGURE 5-22: RATIO OF SO4 DEPOSITION BETWEEN 82-83 AND 80-81

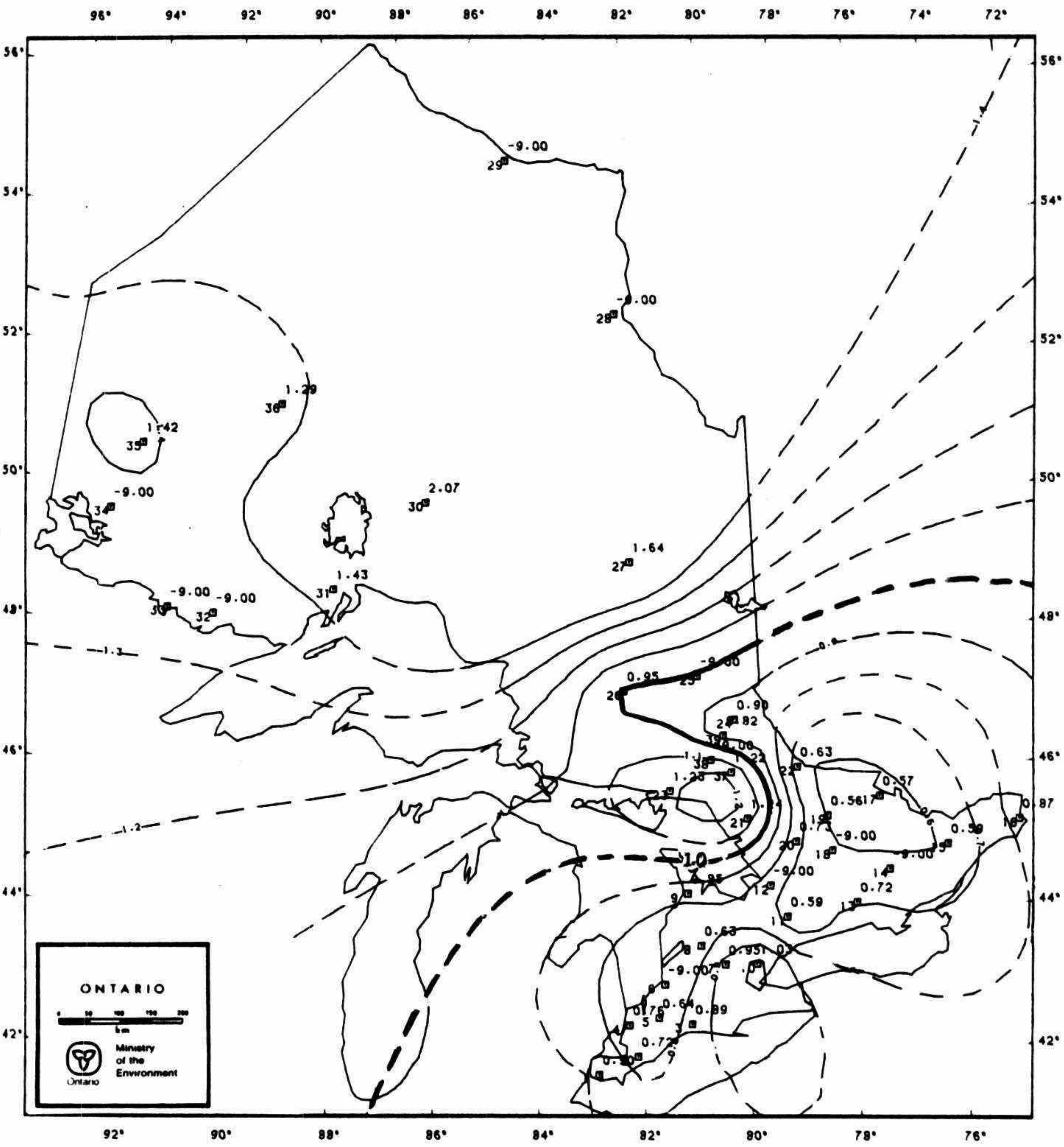


FIGURE 5-23 : RATIO OF SO4 DEPOSITION BETWEEN 82-83 AND 81-82

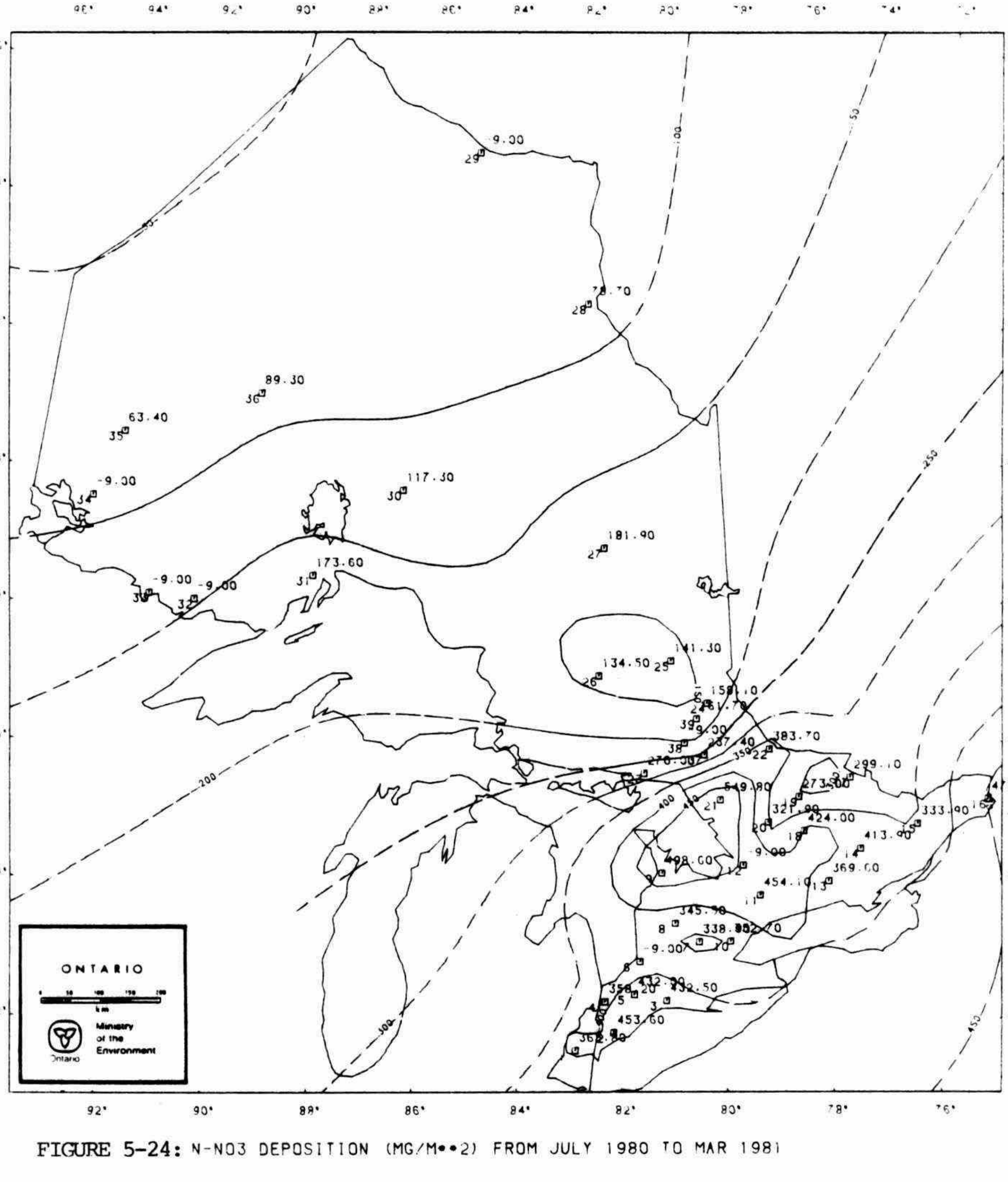


FIGURE 5-24: N-NO3 DEPOSITION (MG/M**2) FROM JULY 1980 TO MAR 1981

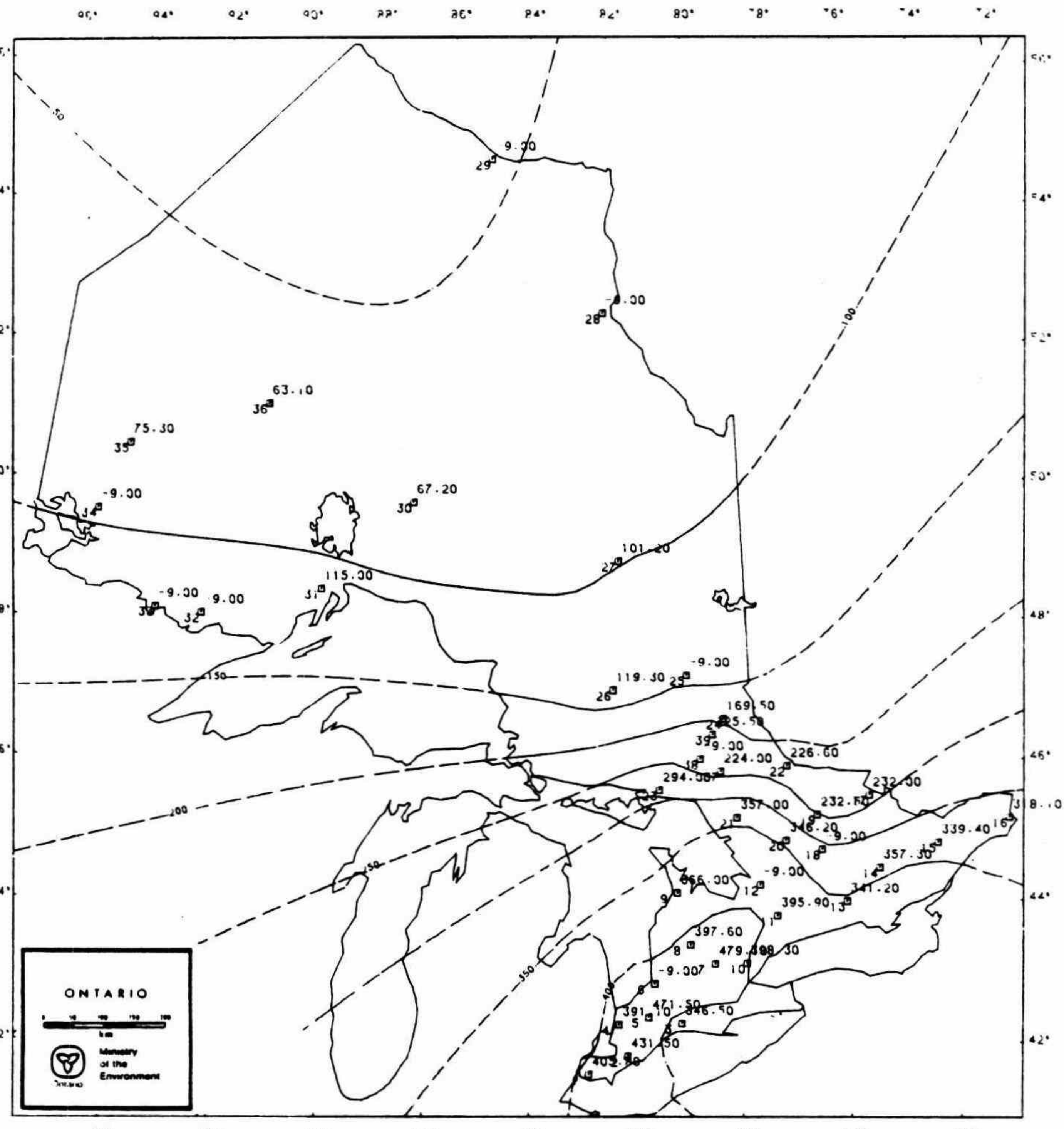


FIGURE 5-25: N-NO3 DEPOSITION (MG/M••2) FROM JULY 1981 TO MAR 1982

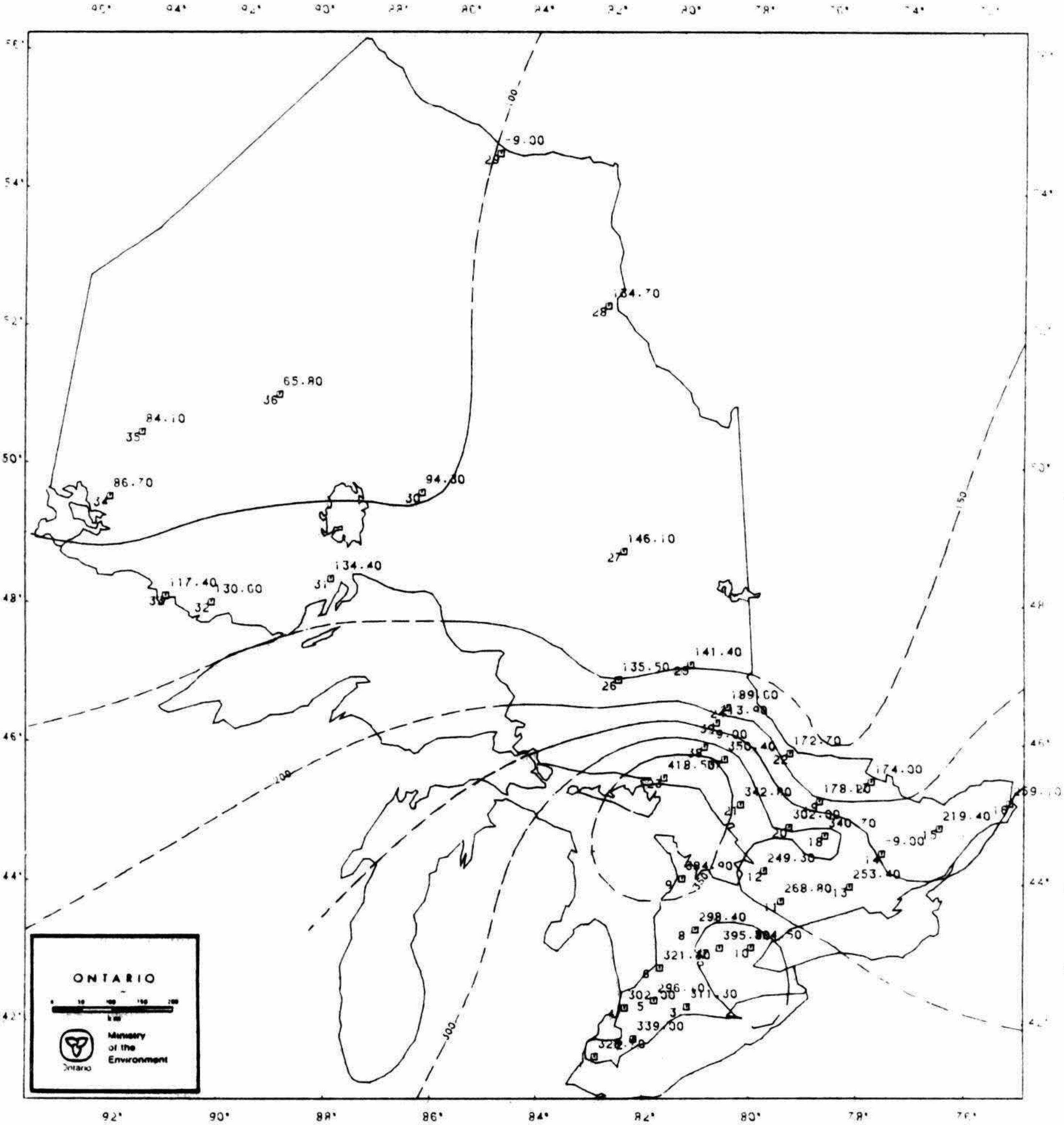


FIGURE 5-26: N-NO3 DEPOSITION (MG/M**2) FROM JULY 1982 TO MAR 1983

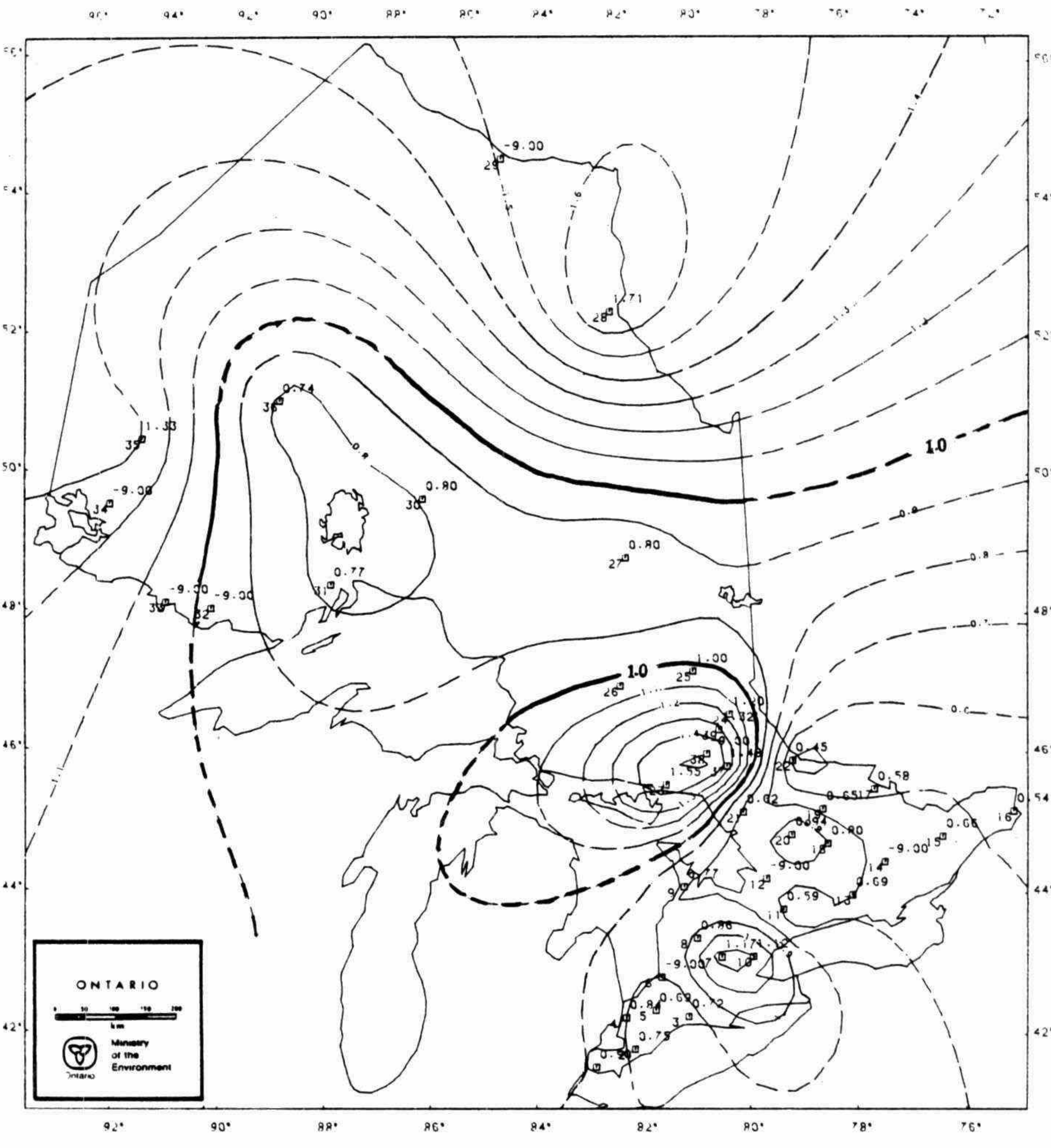


FIGURE 5-27: RATIO OF N-NO3 DEPOSITION BETWEEN 82-83 AND 80-81

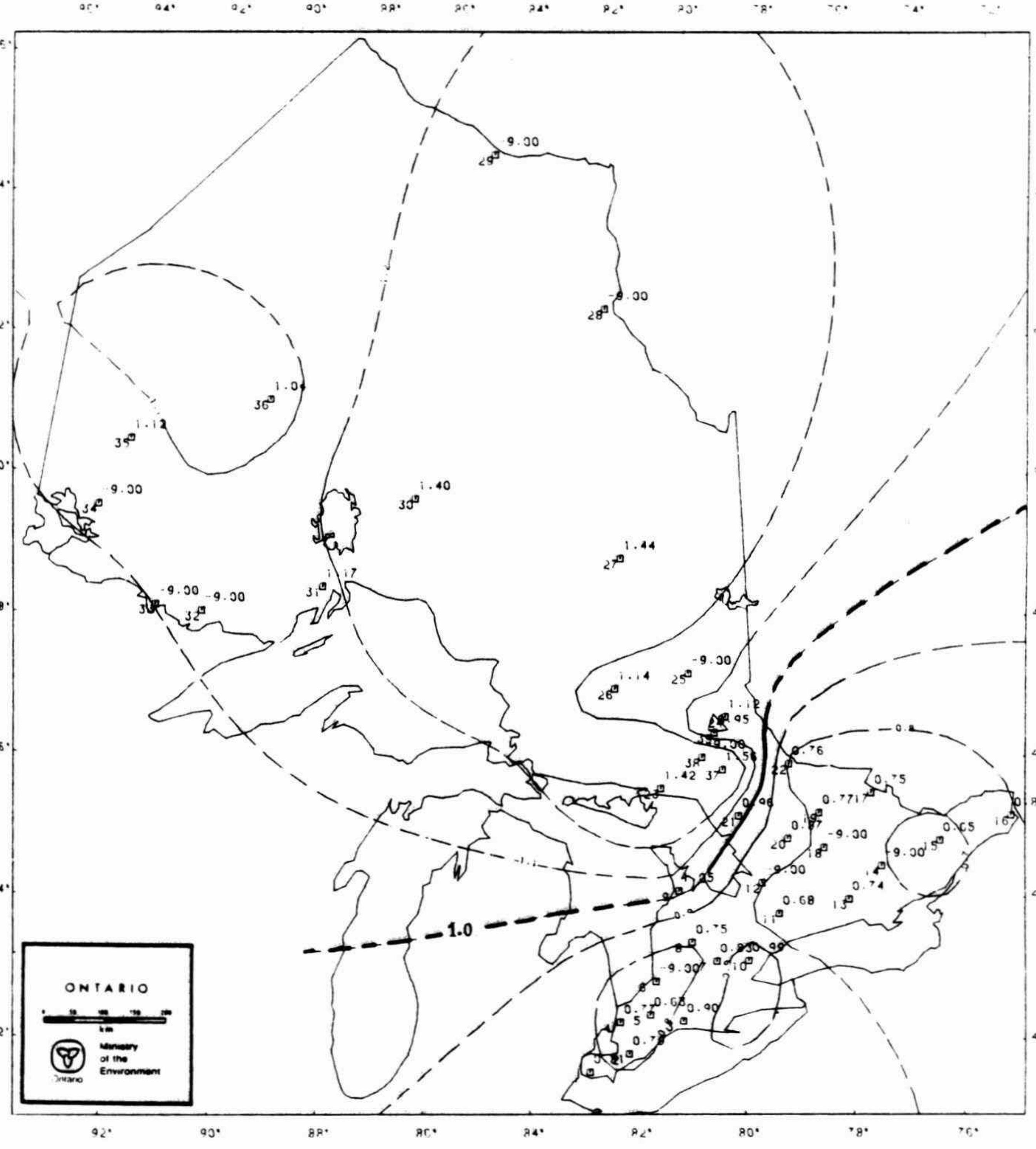
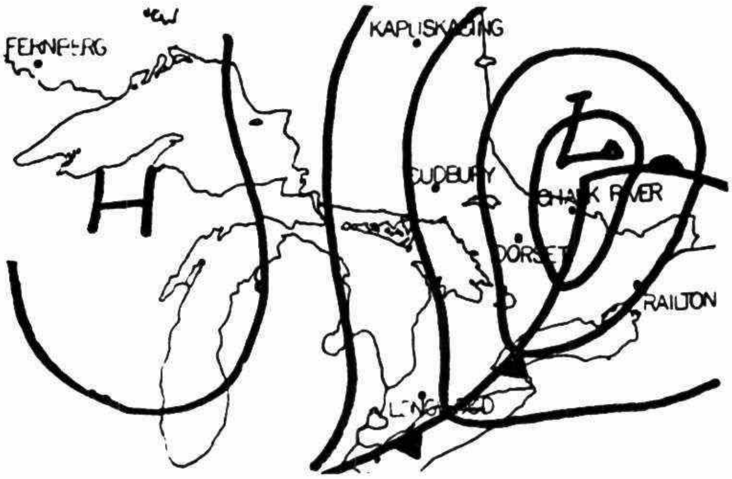
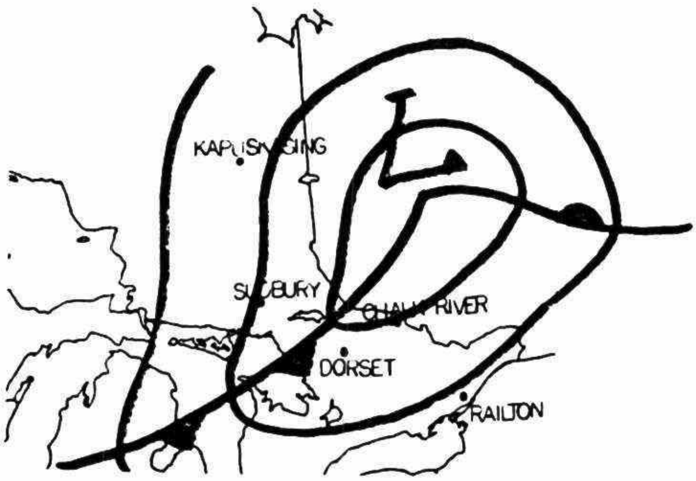


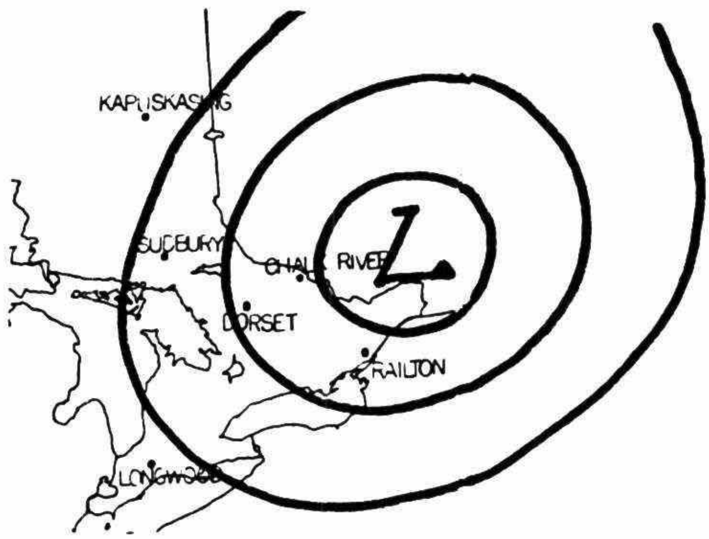
FIGURE 5-28: RATIO OF N-NO3 DEPOSITION BETWEEN 82-83 AND 81-82



1a) Post-cold Front Pre-high Pressure Ridge

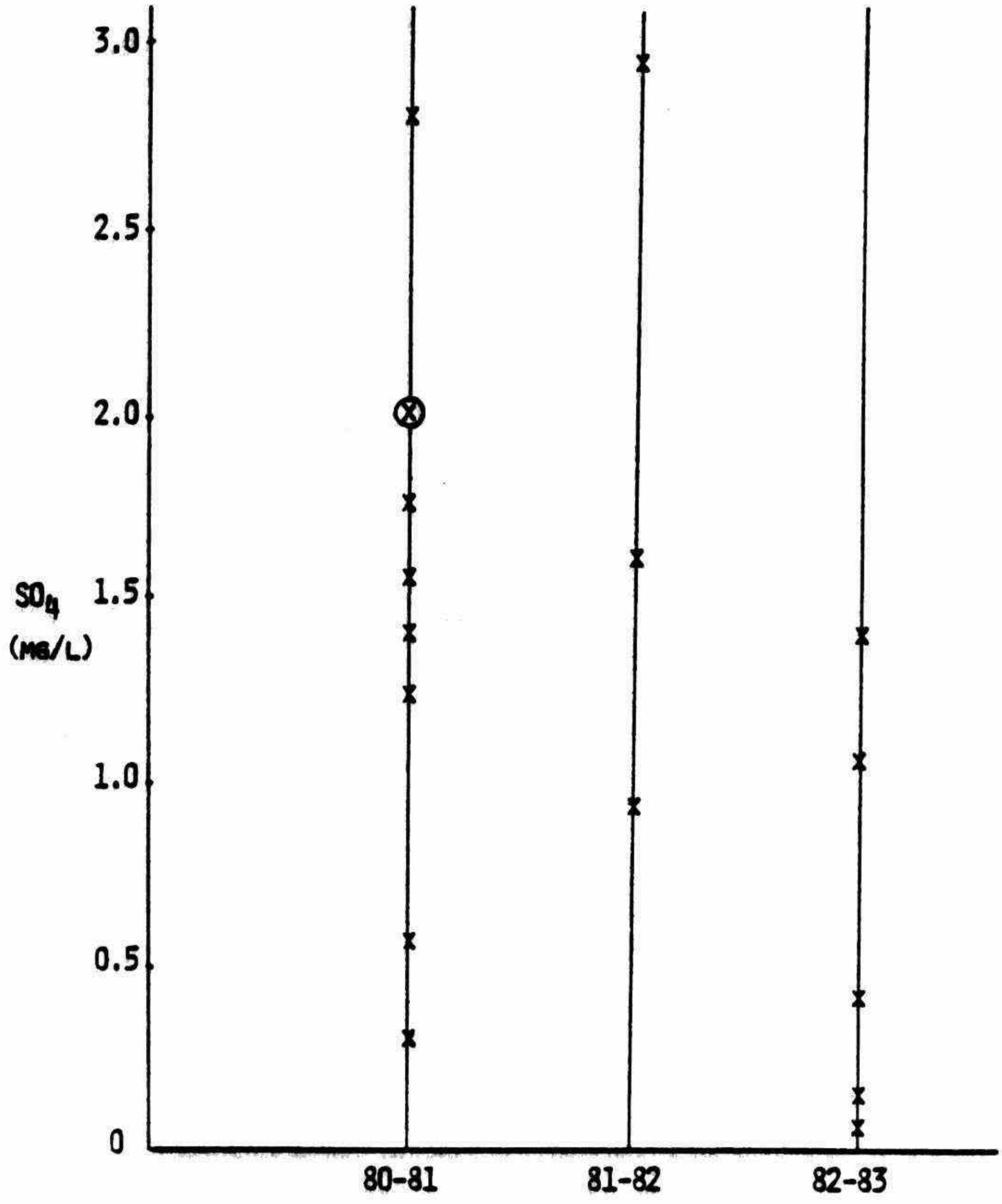


1b) Cold-front Moving Through



1c) Southwest of a Low Pressure System

Figure 6-1. Synoptic Patterns of Possible Sudbury Smelters Contributed Cases.

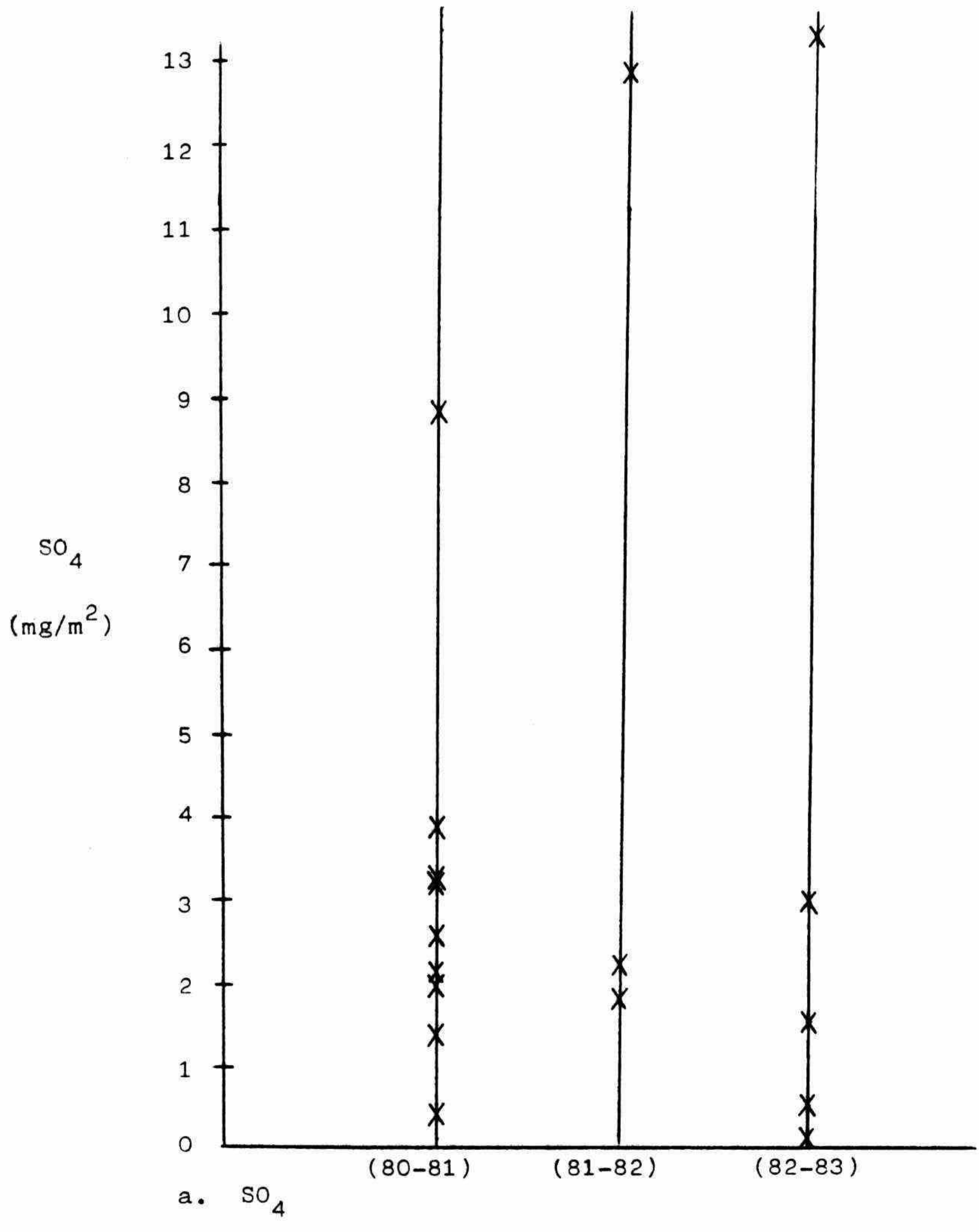


a. SO_4

Figure 6-2. Precipitation Concentration of the Most Possible Sudbury Smelters Affected Cases at Dorset. (⊗ denotes 2 points.)

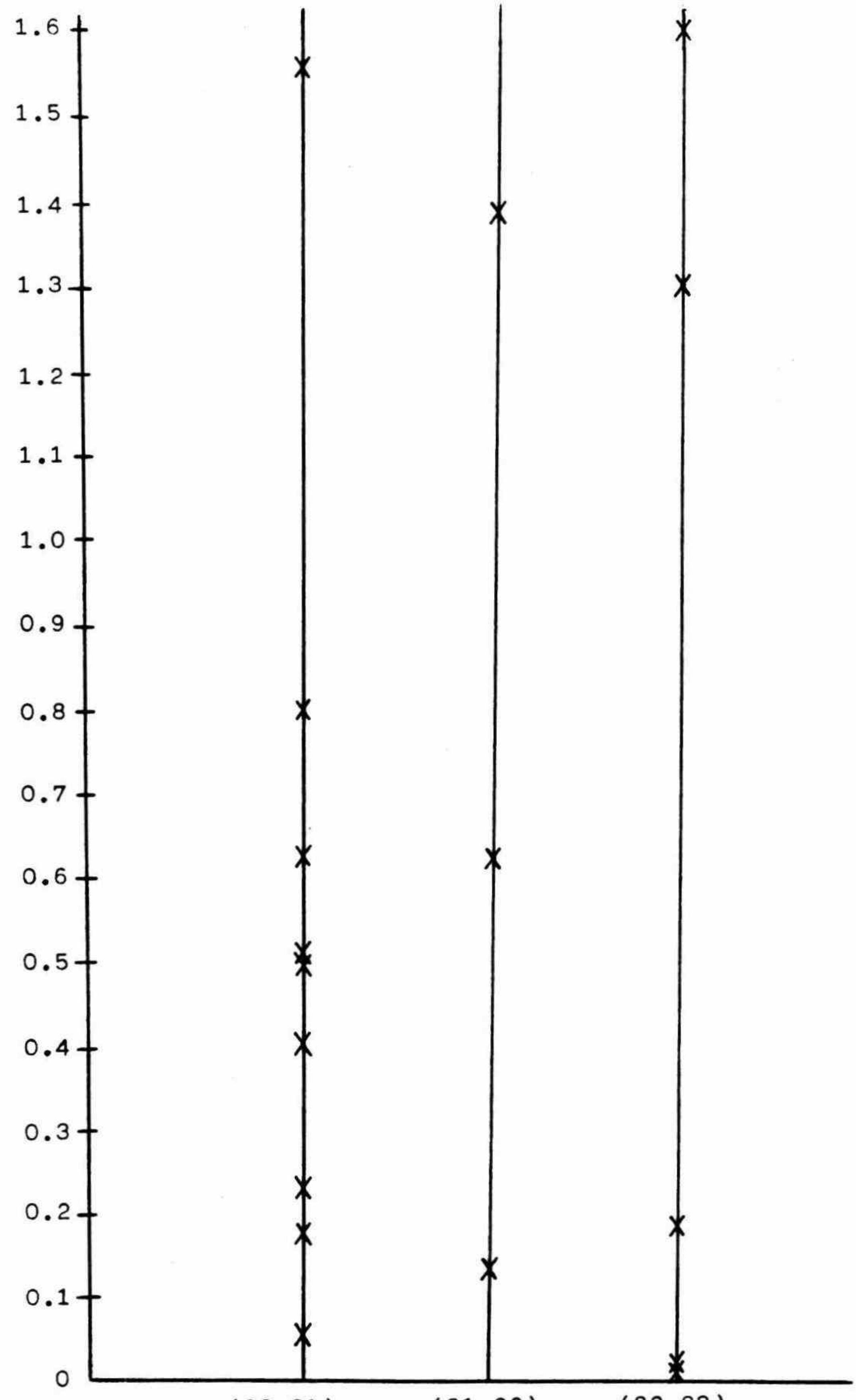
b. N-NO_3

Figure 6-2. (Cont'd)



a. SO_4

Figure 6-3. Deposition of the Most Possible Sudbury Smelters Affected Cases at Dorset



b. $N-NO_3$

Figure 6-3. (Cont'd)

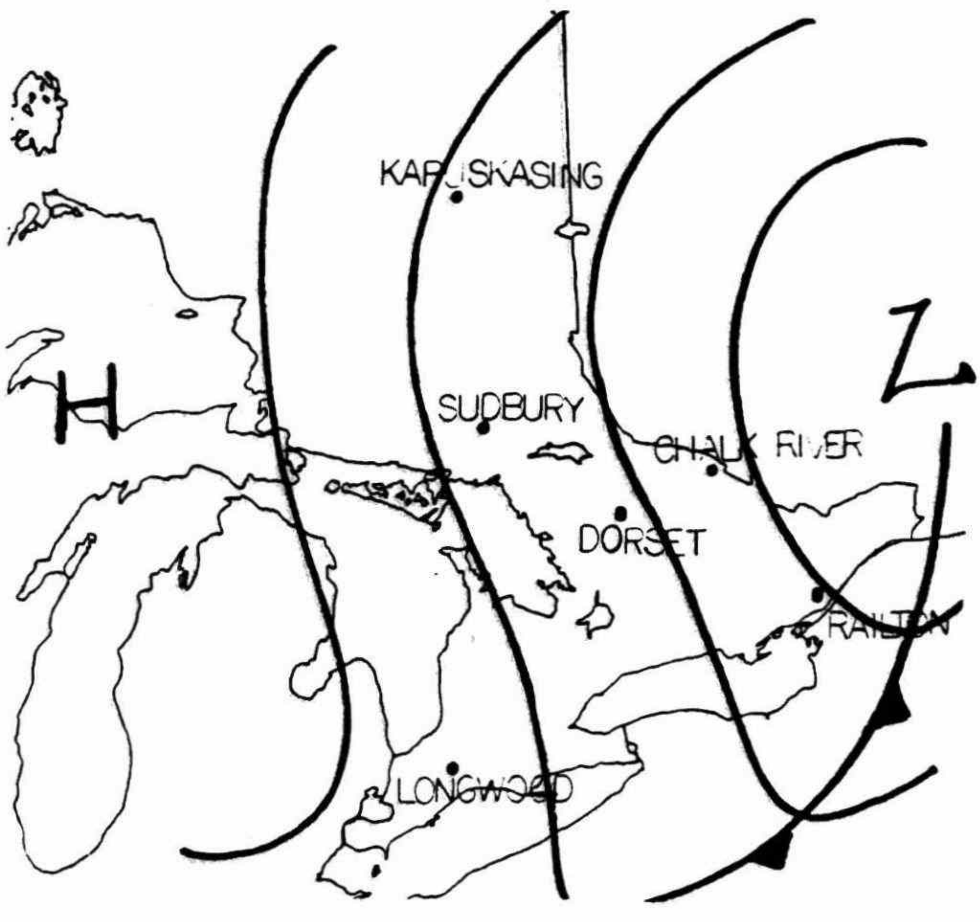


Figure 6-4. Synoptic Pattern of the Most Possible Sudbury Smelters Affected Cases at Railton.

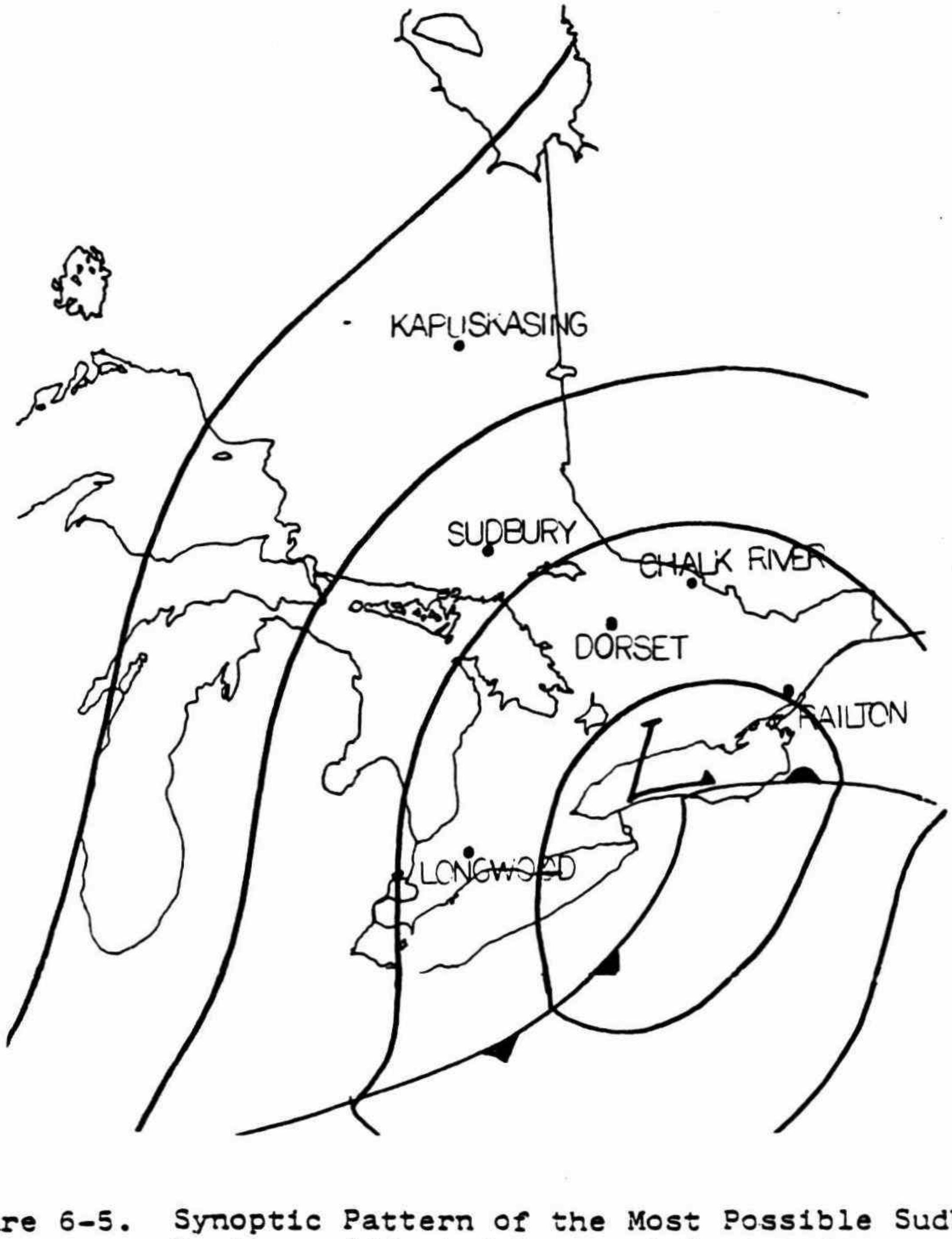
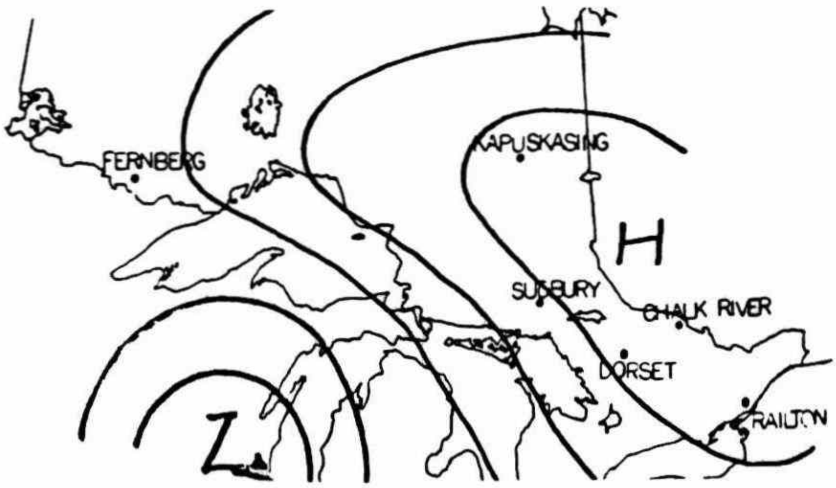
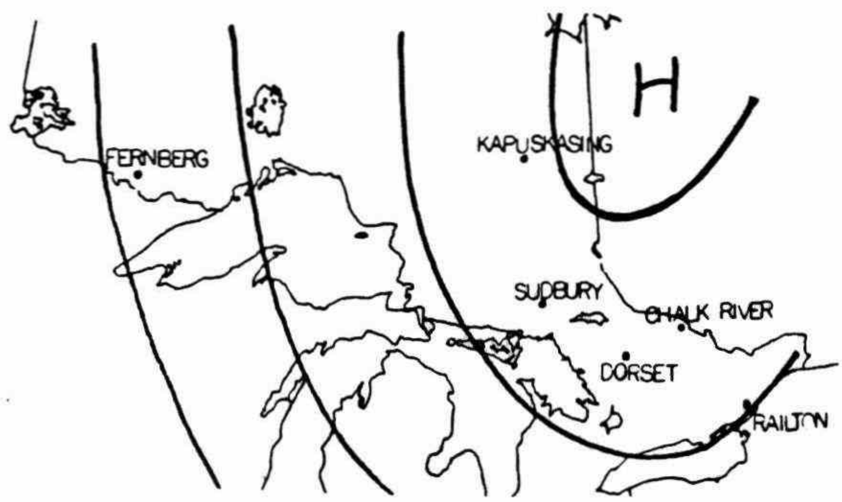


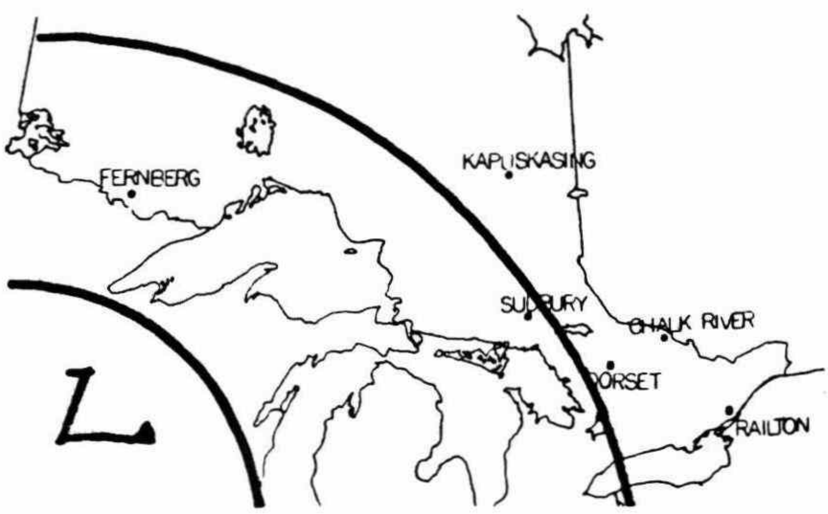
Figure 6-5. Synoptic Pattern of the Most Possible Sudbury Smelters Affected Cases at Longwoods.



a. Northwest of a high pressure system



b. Southwest of a high pressure system



c. Northeast of a Low pressure system

Figure 6-6. Synoptic Patterns of the Most Possible Sudbury Smelters Affected Cases at Fernberg.

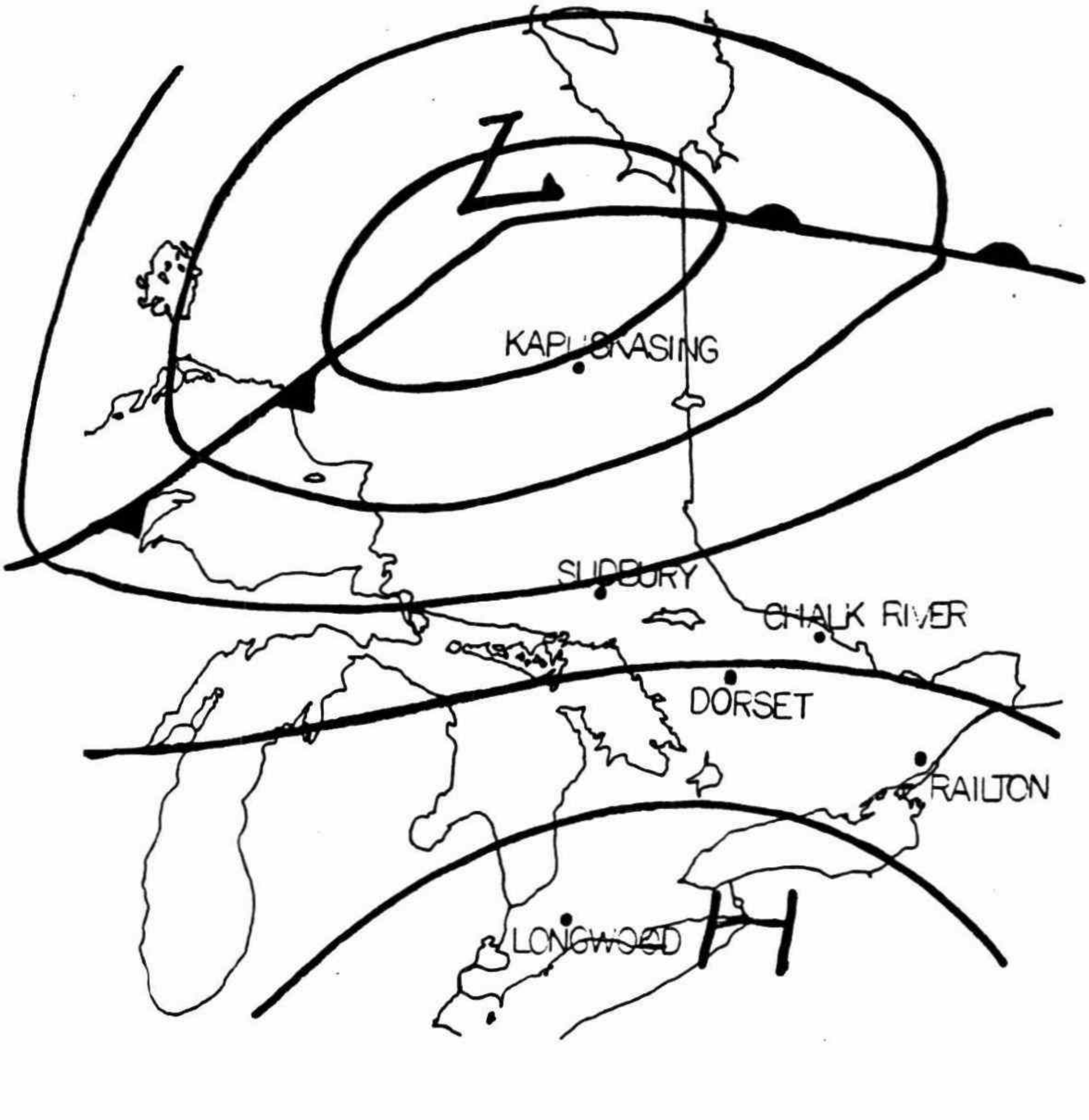


Figure 6-7. Synoptic Pattern of the Most Possible Sudbury Smelters Affected Cases at Chalk River.

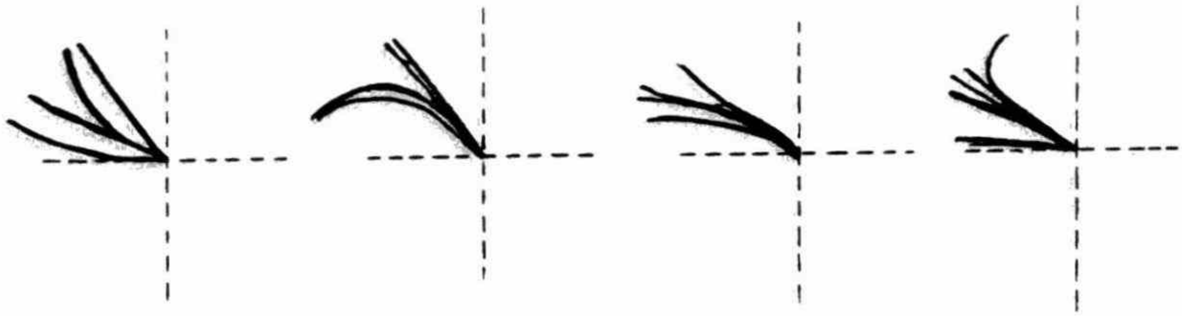


Figure 7-1. 48-hour Back-trajectory Patterns of the Most Possible Sudbury Smelters Affected Cases at Dorset.

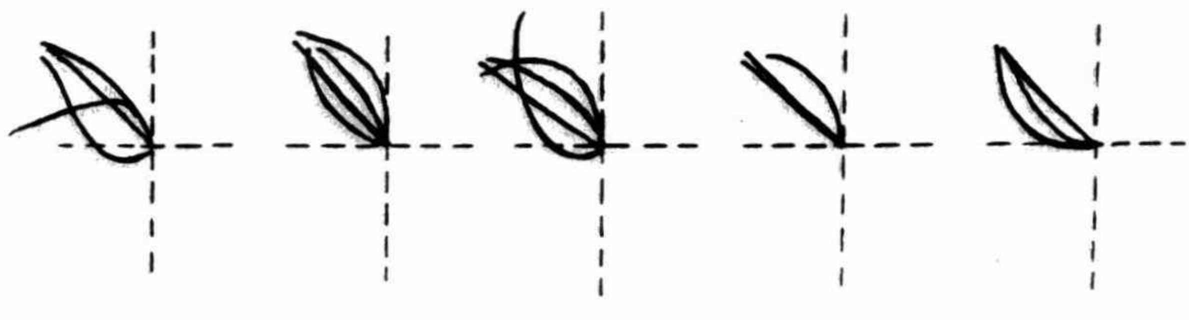
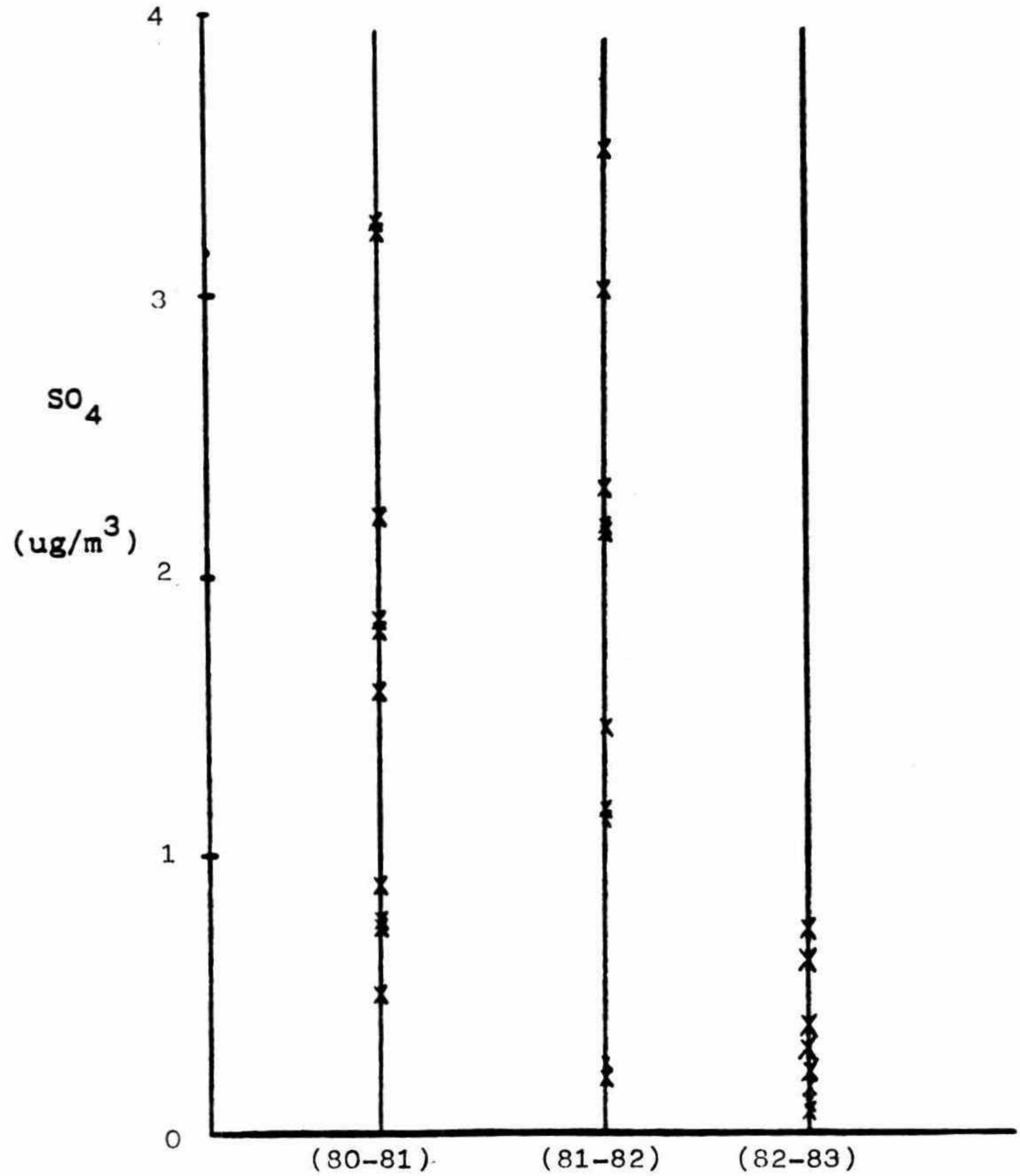
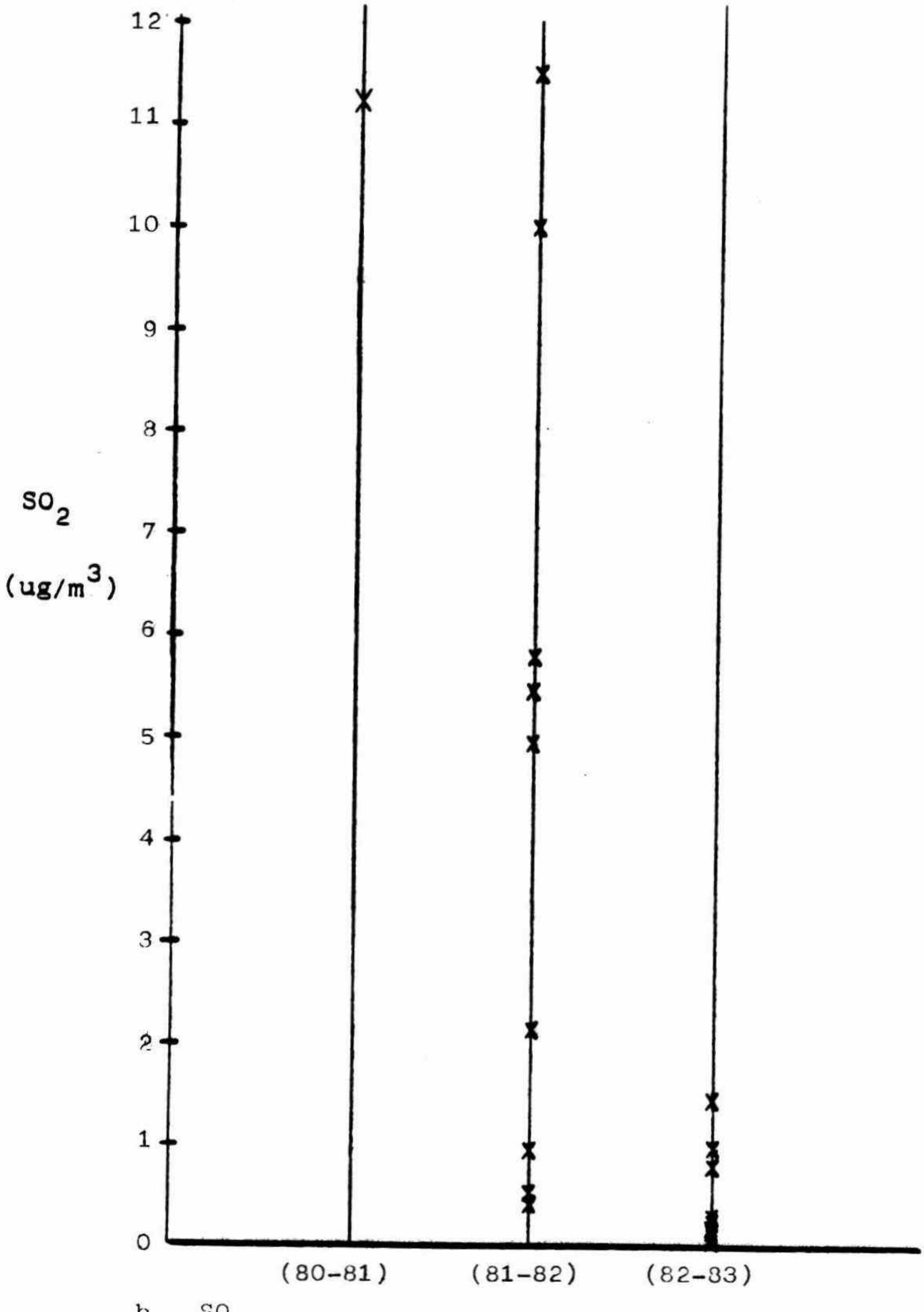


Figure 7-2. 48-hour Back-trajectory Patterns of the Second Most Possible Sudbury Smelters Affected Cases



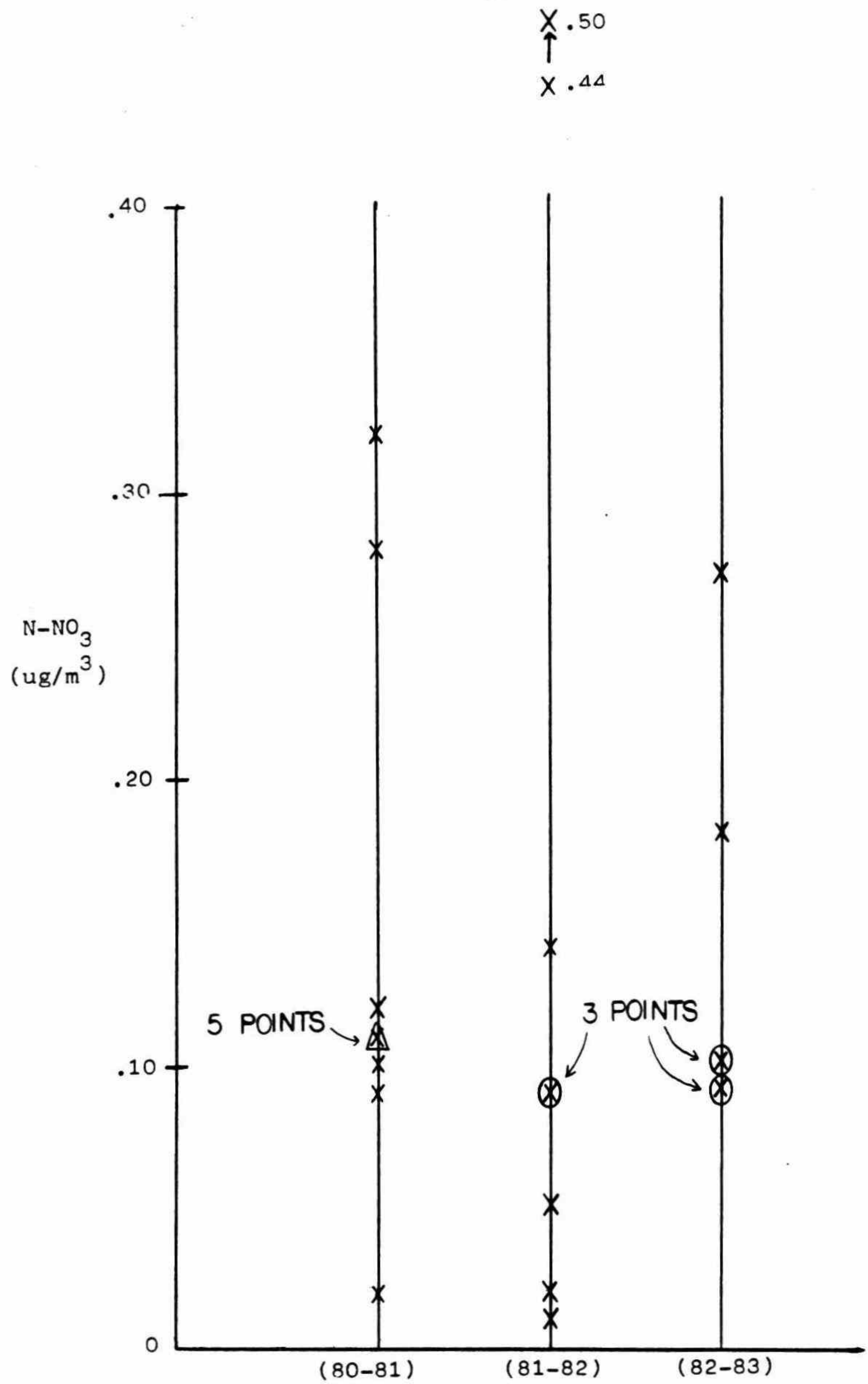
a. SO_4

Figure 7-3. Ground-level Air SO_4 Concentration of the Most Possible Sudbury Smelters Affected Cases at Dorset.



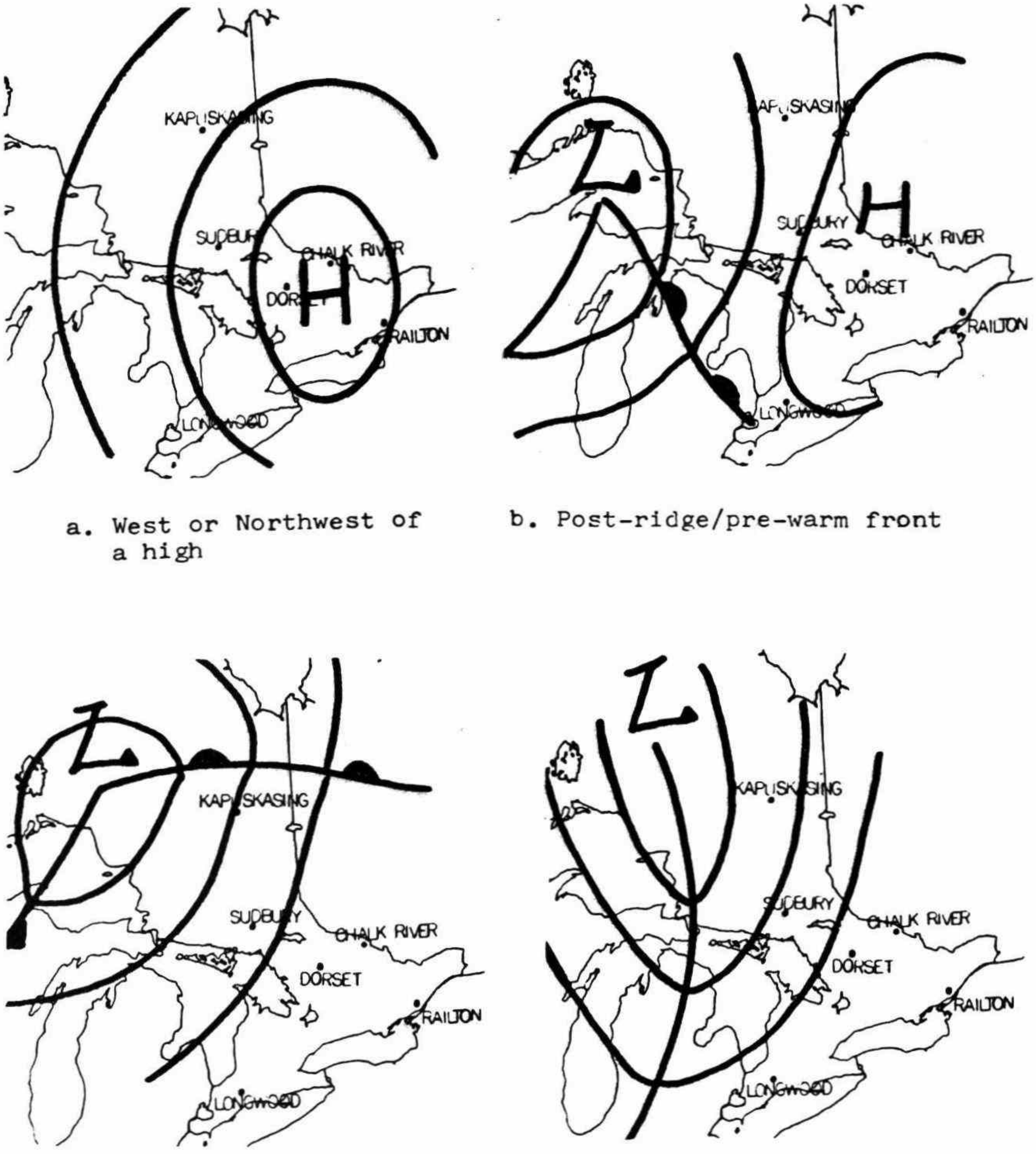
b. SO_2

Figure 7-4. Ground-level Air SO_2 Concentration of the most Possible Sudbury Smelters Affected Cases at Dorset.



c. $N\text{-}NO_3$

Figure 7-5. Ground-level Air NO_3 Concentration of the Most Possible Sudbury Smelters Affected Cases at Dorset.



a. West or Northwest of a high

b. Post-ridge/pre-warm front

c. Warm sector (with southerly flows)

d. Pre-cold front (north-south oriented)

Figure 7.6. Synoptic Patterns of the Most Possible Sudbury Smelters Affected Cases at Kapuskasing. (Receptor area is hatched. Source area is to the S of the receptor.)

APPENDIX 1

Sudbury Shutdown Study Data Stratification Method for Daily Precipitation Samples at the Dorset site

This appendix discusses the data stratification method used for daily precipitation samples, and the related synoptic conditions.

1. Background

Due to the fact that INCO emission sources are not the only important sources and that the samples collected are subject to the effects of complicated meteorological processes, the data have to be properly stratified to take into account the important meteorological processes.

The present stratification scheme attempts to group together data which were obtained under similar (or the same) meteorological (transportation, dilution, stagnation, wet/dry deposition) and chemical (transformation, oxidation) conditions. Traditionally, data stratification is done by stratifying data only according to trajectory sectors, to account for the different source strength in different compass direction. However, this is not sufficient, since a trajectory sector is not associated with a unique synoptic condition. In other words, a simple trajectory analysis does not fully account for the meteorological variability. As a consequence, trajectory sector statistics vary every year even when the emission rates of all sources within a given sector are constant. Besides, a given trajectory sector, unless very carefully selected, may not relate to a unique combination of sources, hence, the stratified data still may have a wide range of variation.

Data are stratified by another frequently used method according to synoptic patterns, since similar patterns are associated with similar meteorological conditions. However, this method cannot easily take the emission sources into account,

since the synoptic pattern changes with time at a specific site. The variation of the affecting sources in a synoptic pattern makes the statistics of a synoptic pattern vary between years, and the data associated with a specific synoptic pattern has a wide range.

For these reasons, the method used here was developed. However, it is first necessary to understand clearly the meteorological situations when precipitation at the receptor is occurring. In the following section, the important meteorological considerations related to data stratification are given.

2. Meteorological Considerations.

Precipitation may be classified as frontal and non-frontal types. For the non-frontal type, the use of single level trajectory (surface or 850 mb level) may be sufficient to distinguish between sources. However, the frontal type needs special considerations.

For trajectory analysis to be useful, the trajectories must correctly indicate the origin of the polluted air masses which have brought about most of the deposition at the receptor. In frontal situations, this may not always be the case, since the flows that move up along the front into the cloud neither follow a constant pressure surface nor a constant height surface (Figure A-1). Surface geostrophic trajectories indicate the correct surface air parcel movement only if the calculated trajectories do not intercept the surface front (since no air parcel moves across the front). In other words, the surface geostrophic trajectories may only be used to estimate the air parcel origin when they are in a cool sector (before warm front), a warm sector (after warm front, before cold front) and in a cold sector (after cold front).

For example, the Sudbury emissions may contribute to Dorset precipitation samples when the samples are collected in a cold sector (this includes three types of patterns: (1) post cold front; (2) cold front passage; (3) precipitation to the

southwest of a low pressure system). In these cases surface geostrophic trajectories may be used to stratify the Dorset precipitation samples, since the Sudbury sources' emissions are transported mainly by cold sector flows (below cold front) to the Dorset region. However, especially in the case of frontal passages, trajectories at different vertical levels have to be examined due to wind shear and the elevated nature of the Sudbury emissions. If an attempt is made to get around this problem by widening the sector for which Sudbury has a potential influence (and using only geostrophic trajectories, for example), then the variability due to meteorological factors is increased. In this present analysis, we looked at the surface geostrophic trajectories and the synoptic weather maps to decrease meteorological variability.

Synoptic patterns have been classified into the following types defined by Heidorn (Water, Air and Soil Pollution, 11, 225-235 (1979)).

(1) post cold front/pre-high pressure ridge

(2) high pressure ridge

(3) post-ridge/pre-warm front

(4) warm front

(5) cyclonic

(6) cold front

(7) stationary front

(8) weak pressure gradient

This classification which was originally developed for a climate study, was here extended for the present study, taking into account extra factors such as stagnation, transporting flow direction and rate, and precipitation location with respect to the synoptic pattern.

This resulted in the following classification scheme (Figure A-2).

1. Post-cold fron/pre-high pressure ridge
   (a) northwest to the low pressure system
   (b) southwest to the low pressure system

2. Warm frontal and warm sector

This type is ususlly associated with a stagnation high pressure system (located in southeast U.S. continent) which moves slowly eastward into the ocean. The southerly flow brings heavily polluted air into the warm frontal clouds. This is a typical episode type synoptic patttern.

3. Post-ridge/pre-warm front

4. Warm front

5. Warm sector

6. Cold front

7. East-west oriented cold front or quasi-stationary front

   (a) north of the front

   (b) south of the front

8. Northwest of a high pressure system

9. Southeast of a high presure system

10. East of a high/warm frontal

11. Cyclone

   (a) NE of cyclone

   (b) NW of cyclone

   (c) SE of cyclone

   (d) SW of cyclone

   (e) centre of cyclone

12. Occluded front

   (a) pre-occluded front

   (b) post-occuluded front

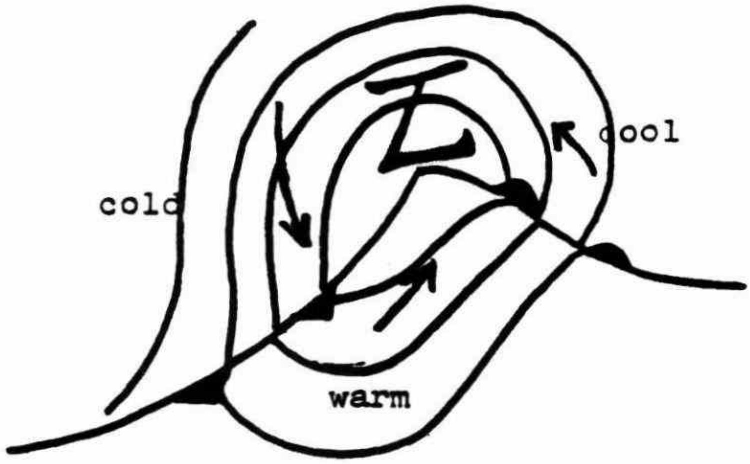
13. Others.

For the precipitation Sudbury impact study, category 1 above was examined in detail with the Dorset data, for example, because it is the situation where there is the highest probability that a Sudbury smelter effect will be seen at Dorset.

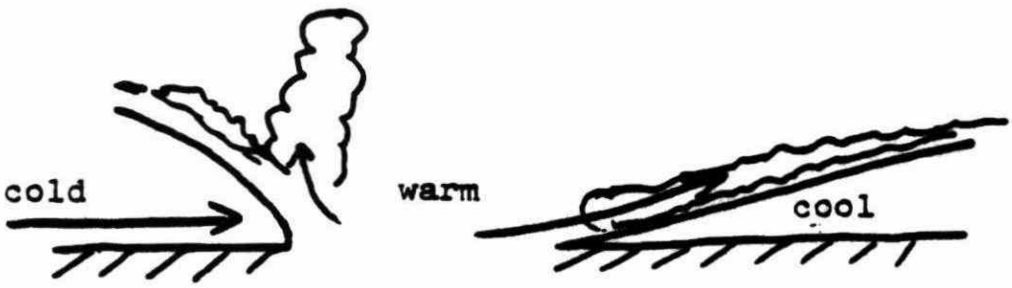
Since the synoptic system is not stationary, it is necessary to classify the sample by all the possible combinations of the above classes. In the shutdown study, samples corresponding to mixed synoptic patterns should not be used in sample comparisons.

From the above considerations, it is clear that we need both trajectory and synoptic pattern to stratify data instead of only stratifying according to trajectory or synoptic pattern alone . Using both trajectory sector and synoptic pattern to stratify the precipitation data, between-year variation of the average sample concentration can be minimized. For example, the average concentration of the class of post-cold front/pre-high ridge class and with trajectories in the NW sector did not change significantly between years if the strength of the emission sources remain constant.

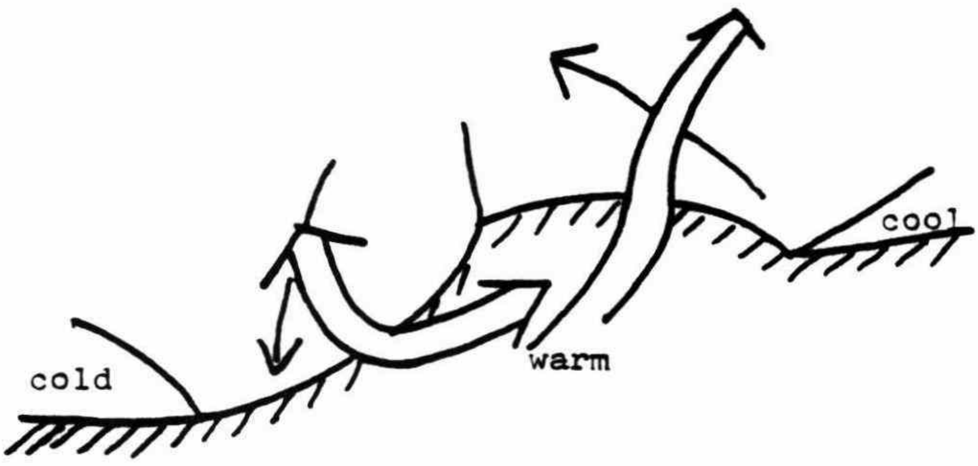
It has to be mentioned here that the use of trajectory sector and synoptic pattern to stratify data does not fully account for meteorological variability, since the above synoptic pattern classification does not fully account for the storm track or geometric shape of the synoptic pattern. The use of a trajectory sector cannot overcome this problem, since trajectories can have different patterns within a sector. In other words, the data after being stratified by trajectory sector and synoptic pattern, still may show the within group variation; however, the between group mean variation is minimized. If the sample size is large enough, we may stratify data by using trajectory pattern and synoptic pattern instead of using trajectory sector. This can minimize the within group data range and provide more specific statistics.



a. Flow on horizontal plane.

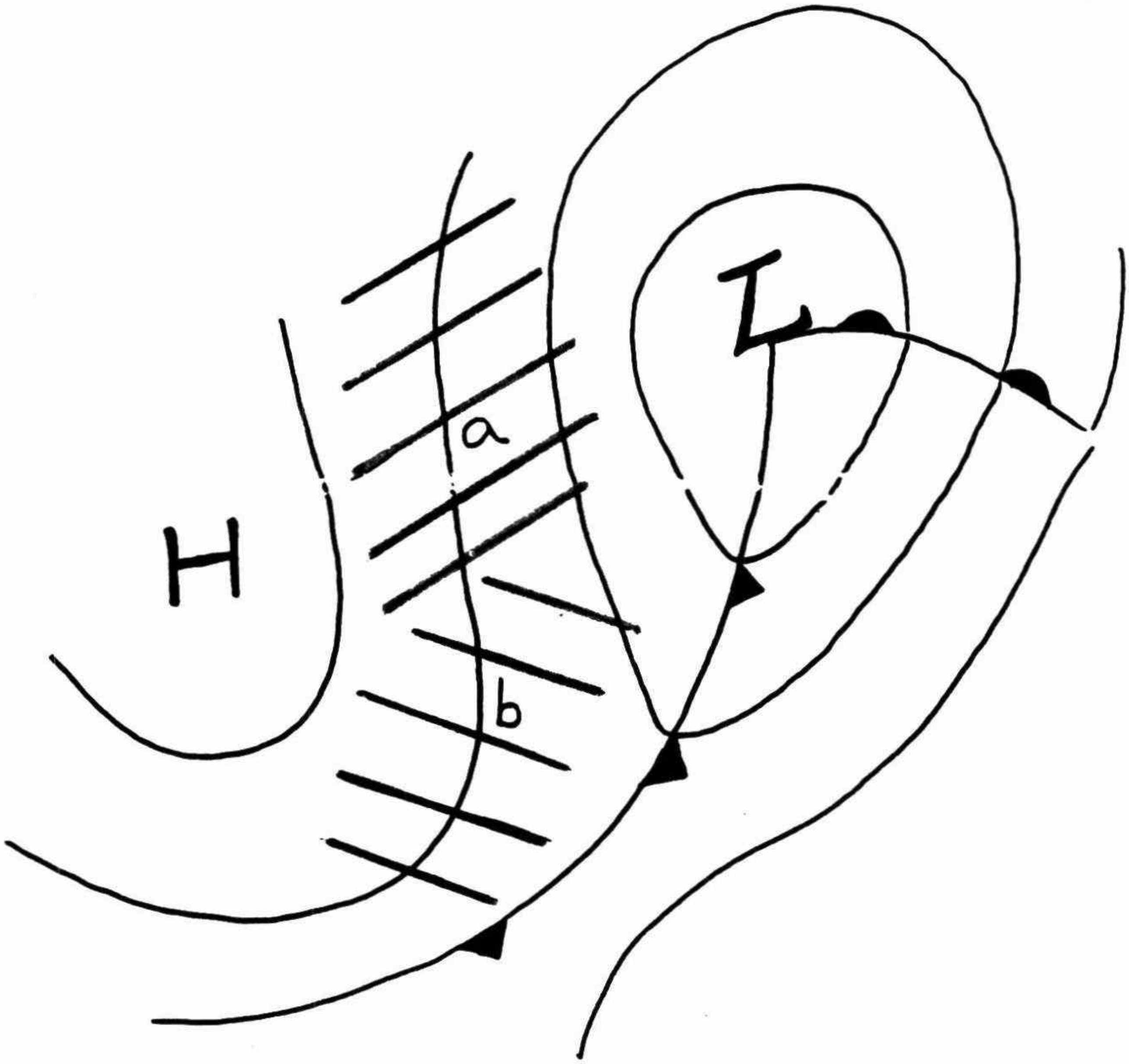


b. Vertical cross section.



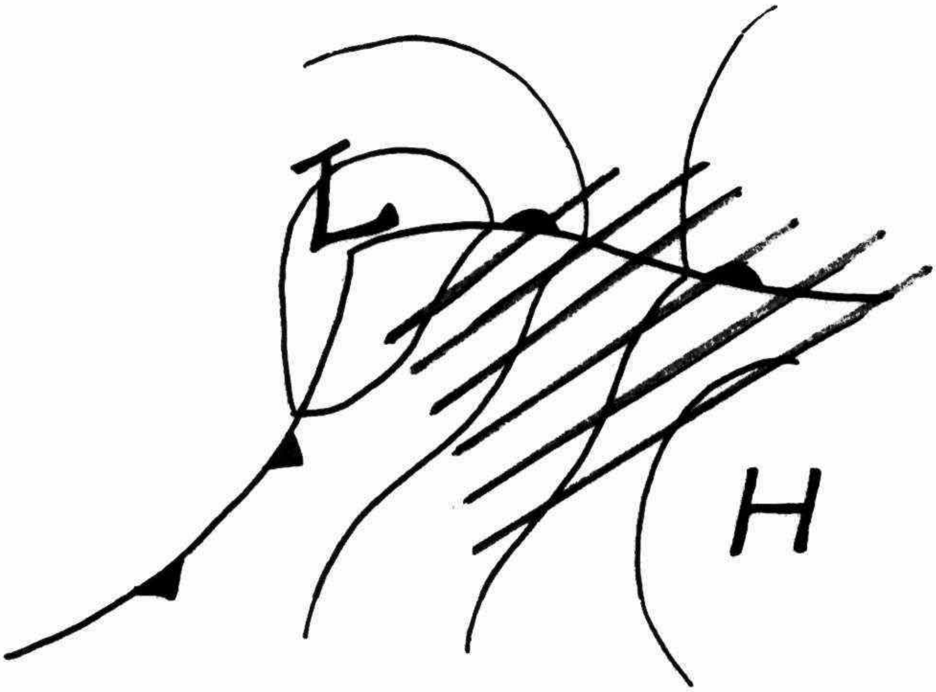
c. 3 dimensional

Figure A-1 :Flow structure at frontal zone.

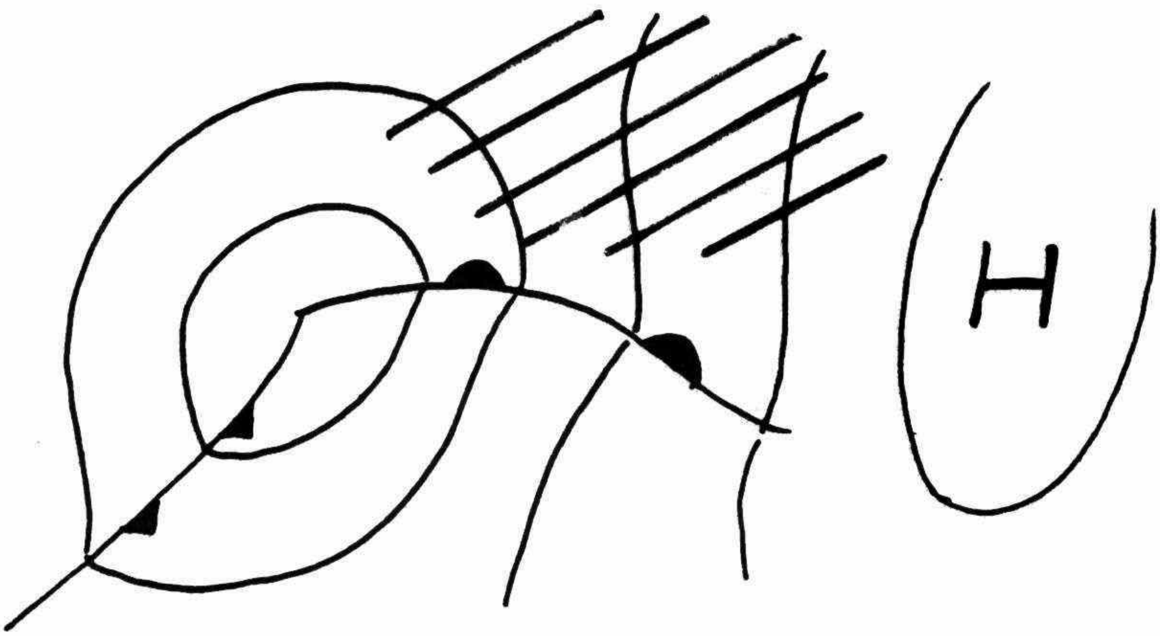


Class 1. Post-cold front/pre-high pressure ridge.

Figure A-2: Synoptic pattern classes.
(Hatched area denotes receptor region.)

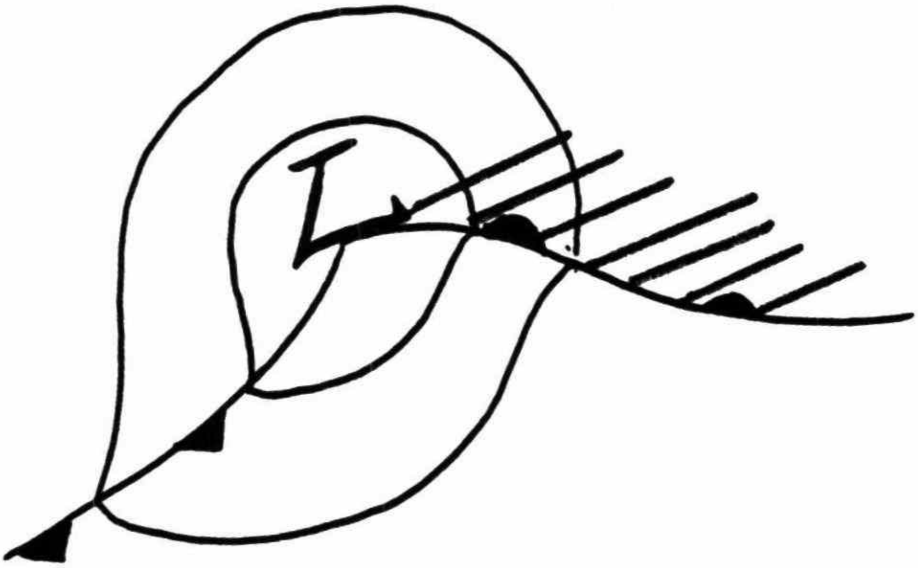


Class 2. Warm frontal and warm sector.

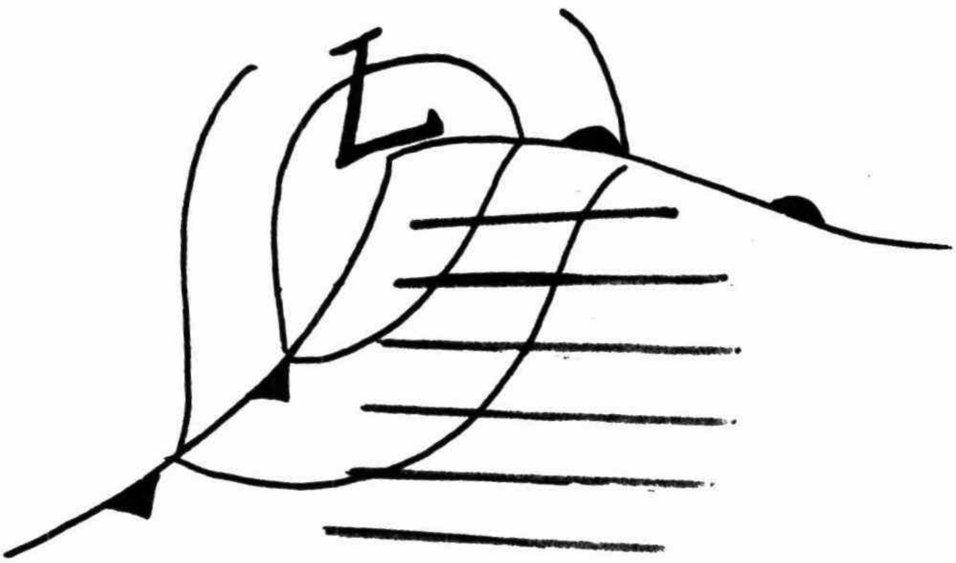


Class 3. post-ridge/pre-warm front.

Figure A-2: (Cont'd).



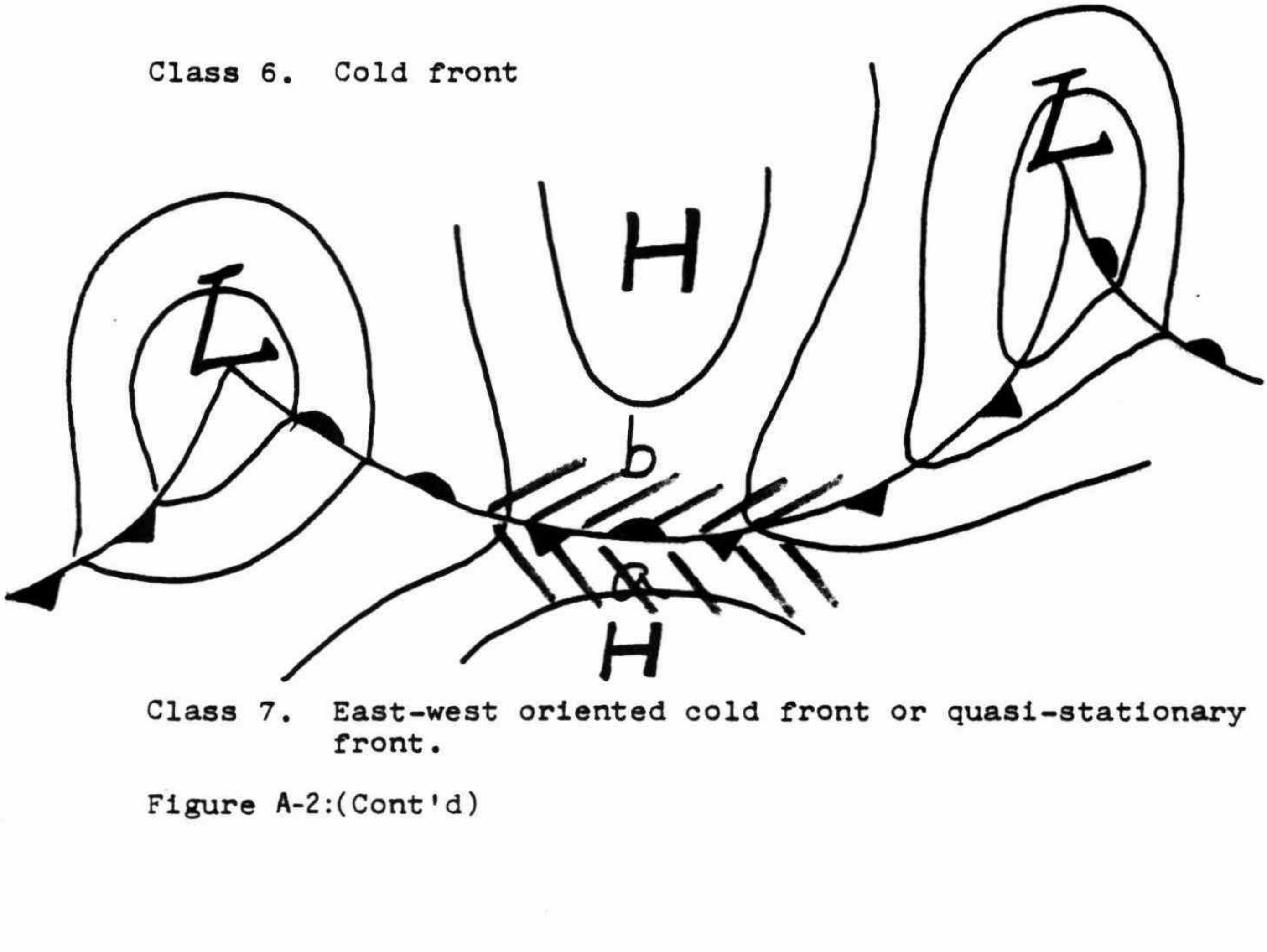
Class 4. Warm front.



Class 5. Warm sector.

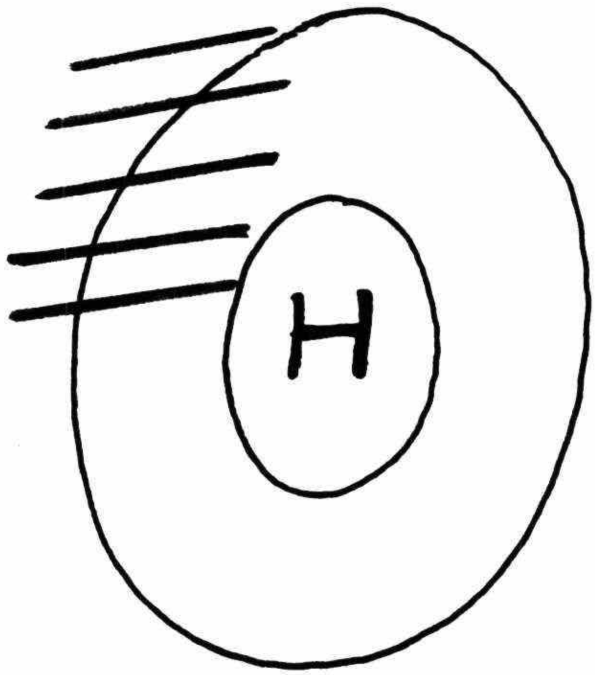
Figure A-2:(Cont'd)

Class 6. Cold front

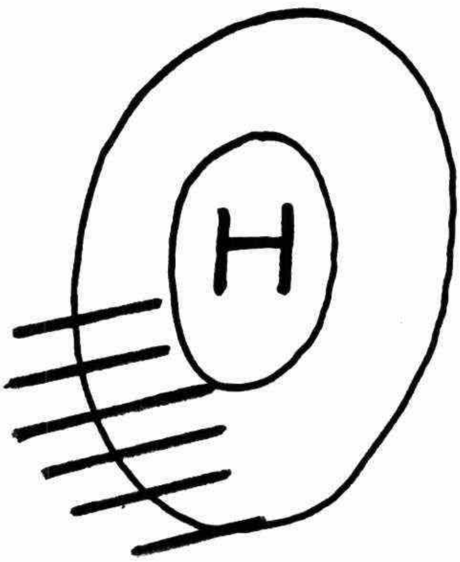


Class 7. East-west oriented cold front or quasi-stationary front.

Figure A-2:(Cont'd)

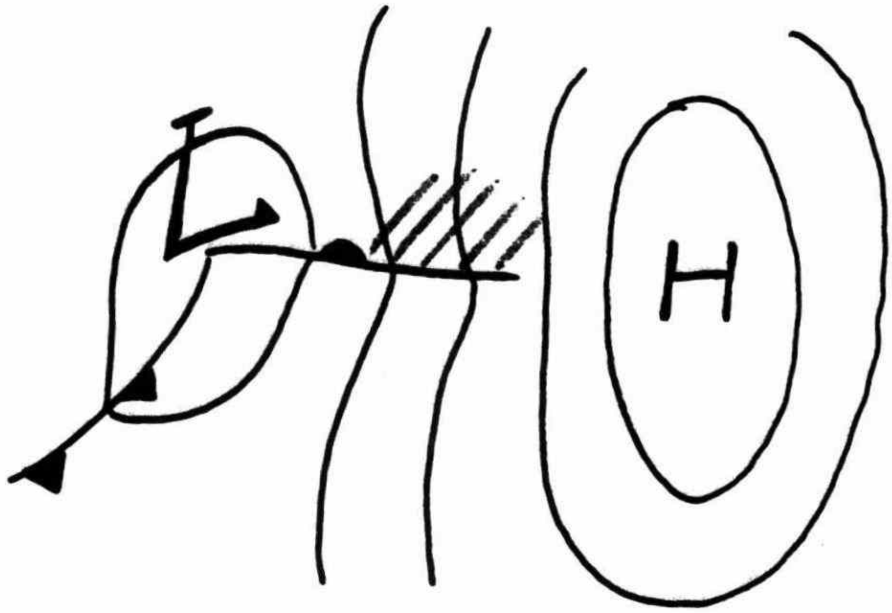


Class 8. Northwest of a high pressure system.

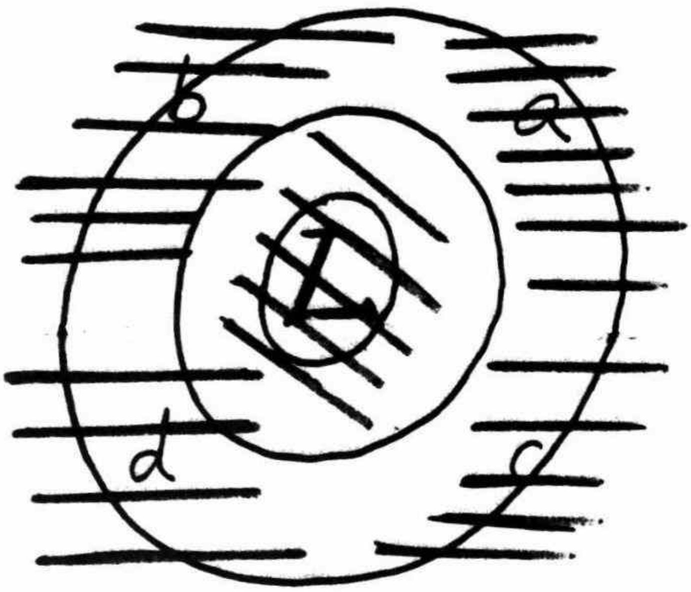


Class 9. Southeast of a high pressure system.

Figure A-2 (Cont'd)

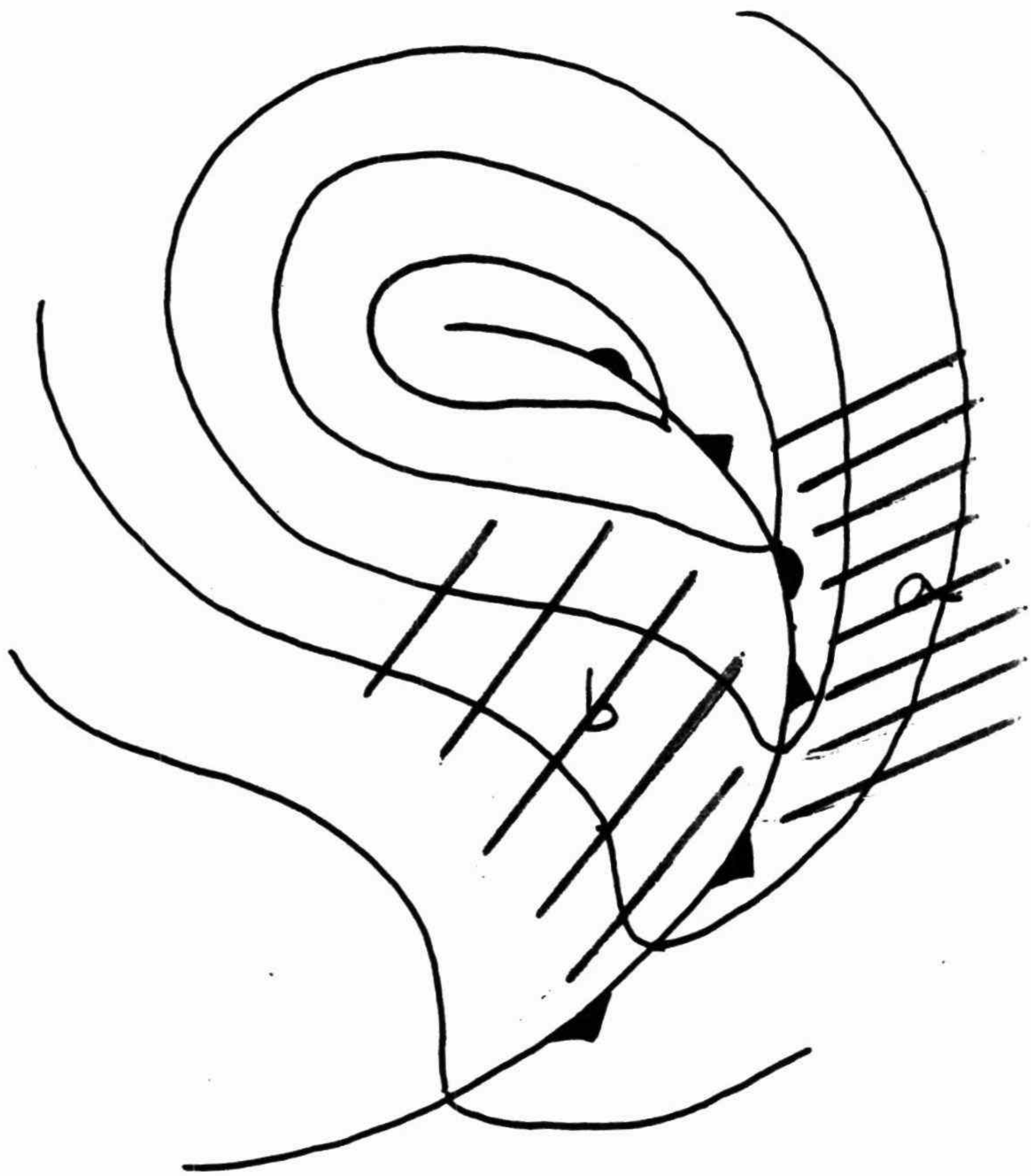


Class 10. East of a high pressure system/warm front.



Class 11. Cyclone

Figure A-2 (Cont'd)



Class 12. Occluded front.

Figure A-2 (Cont'd)